Experimental Investigation of the Stabilization and Structure of Turbulent Cool Diffusion Flames

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Although recent studies of laminar cool flames have provided important advances in understanding the low-temperature chemistry of both hydrocarbons and oxygenates, there has been limited experimental insight into how interactions between turbulence and chemistry occur in cool flames. To address this, a new Co-flow Axisymmetric Reactor-Assisted Turbulent (CARAT) burner has been developed and characterized in this investigation for the purpose of directly studying turbulent cool flames. A methodology for establishing stable turbulent cool diffusion flames under well-defined conditions is proposed. The structure of dimethyl ether flames is examined using both formaldehyde planar laser-induced fluorescence and Rayleigh scattering. It is found that weak turbulence produces wrinkled turbulent cool flames in which fluctuations occur mainly on the fuel side of the flame. However, at increased levels of turbulence, large pockets of unburned reactants appear in the vicinity of the cool flame, and structural fluctuations extend to both sides of the flame. This study offers a well-defined experimental platform for the study of turbulence-chemistry interactions at low temperatures.

I. Introduction

OVER the past several years, experimental investigations of cool flame phenomena have proliferated. Cool flames have been studied under well-defined conditions in microgravity droplets\textsuperscript{1,2}, counterflow burners\textsuperscript{3-5}, micro flow reactors\textsuperscript{6}, and Hencken burners\textsuperscript{7}. The results of these studies have shown that the low-temperature chain-branching sequence (peroxy chemistry) present in many hydrocarbons and oxygenates is capable of producing enough radicals and heat release to sustain stable cool flames across a variety of conditions. These experiments have also provided important validation targets for the development of chemical kinetic models at low temperatures\textsuperscript{8}.

However, despite the ubiquity of turbulent combustion in real engines, well-defined turbulent cool flame experiments have been extremely limited in number. Gokalp\textsuperscript{9} and Koliatis\textsuperscript{10} studied premixed cool flames within reactors subject to various degrees of turbulence. Some of the authors’ previous experiments\textsuperscript{11,12} involved turbulent premixed flames that were strongly affected by low-temperature ignition, but no cool flame fronts were actually visible in those experiments. Recent numerical simulations have shown that cool flames can play an important role in the ignition\textsuperscript{13,14}, stabilization\textsuperscript{15}, and propagation\textsuperscript{16} of turbulent flames. However, numerical prediction of cool flame behavior is extremely sensitive to the chemical kinetic model employed, and many widely used models that

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perform well for high-temperature flames are incapable of describing cool flames accurately\textsuperscript{8}. Therefore, the experimental measurement of turbulent cool flames can provide valuable insights into the modeling of turbulence-chemistry interactions at low temperatures.

In the present study, a new Co-flow Axisymmetric Reactor-Assisted Turbulent (CARAT) burner is employed for the investigation of turbulent cool diffusion flames. Dimethyl ether (DME) is chosen as the fuel due to its relatively strong low-temperature chemistry and known Rayleigh scattering cross-section\textsuperscript{18}. The two-dimensional turbulent cool diffusion flame structure is examined through the formaldehyde planar laser-induced fluorescence technique, while one-dimensional radial temperature profiles are produced using Rayleigh scattering. The effects of increases in turbulence on the cool diffusion flame structure are also discussed.

II. Experiment

The Co-flow Axisymmetric Reactor-Assisted Turbulent (CARAT) burner, depicted in Figure 1, is a generational update to the Reactor-Assisted Turbulent Slot (RATS) burner\textsuperscript{11, 12}. Like the RATS burner, the CARAT burner is a Bunsen-type burner with an extended reactor section capable of controlling the pre-flame residence time and temperature. Two grids located in the reactor section generate turbulence within the main flow. The burner main exit is 15 mm in diameter ($D$). A small annulus (2-mm gap size) surrounding the main exit enables the use of a pilot flow. The CARAT burner also includes a large exterior channel that enables a heated or vitiated co-flow, similar to the well-known Cabra burner\textsuperscript{19}. The co-flow passes through a ceramic honeycomb mesh, which allows for flow laminarization and well-defined boundary conditions.

The turbulence in the main flow has been characterized by a nanoscale thermal anemometry probe (NSTAP)\textsuperscript{20} in combination with a Dantec Dynamics Streamware Pro Constant Temperature Anemometry system. Due to its small size (60 μm long, 2 μm wide, 100 nm thick), the NSTAP is capable of extremely high spatial and temporal resolution. The NSTAP was positioned at the centerline of the main exit and operated at a sampling frequency of 250 kHz. Figure 2 shows the range of turbulence intensities ($u'/U$) and integral length scales ($l$) measured by the probe as a function of the bulk Reynolds number ($Re_D = UD/\nu$). The integral time scales (and therefore the integral length scales) were determined by extrapolating the power spectrum to zero frequency. Since the turbulent flow field characterization was performed at room temperature (295 K), the effects of elevated temperatures on $u'/U$ and $l$ are accounted for by modifying the mixture viscosity appropriately to determine $Re_D(T)$.

To visualize the turbulent cool flame structure, formaldehyde (CH$_2$O) planar laser-induced fluorescence (PLIF) is used. The third harmonic (355 nm) of an Nd:YAG laser (Quantel, Q-smart 850) excites the CH$_2$O at an energy of approximately 200 mJ/pulse. The laser operates at a frequency of 5 Hz. Two band filters, passing frequencies between 400 and 450 nm, are positioned in front of an ICCD camera (Princeton Instruments, PI-MAX 4) to isolate the CH$_2$O fluorescence. The laser beam is expanded into an approximately 200-μm-thick vertical sheet and positioned at the centerline of the burner.

Rayleigh scattering is performed in order to quantify the turbulent cool flame temperature. The second harmonic (532 nm) of a 10-Hz Nd:YAG laser (Spectra-Physics, lab-170-10) is directed through the flame at an energy of approximately 400 mJ/pulse. Accounting for changes in the Rayleigh scattering cross-section within the flame is often the most challenging aspect of the technique\textsuperscript{18}. However, the cool flame’s tendency to exhibit massive fuel leakage eases this difficulty. Using the example of a 1-D opposed-flow flame\textsuperscript{21, 22}, it can be seen in Fig. 3 that an

![Fig. 1. Photograph of the Co-flow Axisymmetric Reactor-Assisted Turbulent (CARAT) burner used in this study.](image1)

![Fig. 2. Results of the CARAT burner turbulent flow field characterization in terms of the turbulence intensity ($u'/U$) and integral length scale ($l$).](image2)
unburning mixture with boundary temperatures of \( T = 500 \) K and a cool flame with boundary temperatures of \( T = 600 \) K have nearly the same mixture properties. The presence of a hot flame, on the other hand, causes significant changes which must be accounted for through extensive corrections of the Rayleigh signal. Therefore, in this study, the cool flame temperature is determined by directly comparing its Rayleigh scattering signal to an unburning flow field with identical mixture properties at a known temperature.

III. Stabilization of Turbulent Cool Diffusion Flames

In this study, turbulent cool diffusion flames are established using dimethyl ether (DME) as the fuel. Under the conditions given in Fig. 4, cool flames are able to be stabilized in the recirculation zone between the main flow (DME/N\(_2\)) and the pilot flow (O\(_2\)). The range of conditions for which a cool flame can be stabilized is highly temperature dependent. Specifically, DME turbulent cool diffusion flames were seen to form readily for \( T = 600 \) K boundary temperatures but not for \( T = 550 \) K or \( T = 500 \) K. The use of pure oxygen in the pilot flow extends the range of conditions for which turbulent cool diffusion flames can be stabilized but is not strictly necessary, as cool flames were also able to be formed with air in the pilot flow. Notably, increasing the main flow velocity (\( U \)) or decreasing the DME mole fraction (\( X_{\text{DME}} \)) does not result in a sharp transition between stable turbulent cool diffusion flames and blowoff. Rather, it was seen that a range of conditions exists for which lifted turbulent cool diffusion flames result, some of which transiently reach blowout over the course of a several minutes. For all measurements in this study, the pilot flow velocity is 1.0 m/s, and the co-flow velocity is approximately 0.55 m/s.

IV. Flame Structure Measurements through CH\(_2\)O PLIF

Due to its unique chemistry, DME is an extremely attractive fuel for the CH\(_2\)O PLIF technique. Multiple reaction pathways are available for DME to decompose into CH\(_2\)O, and as a result the signal-to-noise ratio for CH\(_2\)O PLIF of DME flames is typically quite high. DME cool flames offer even stronger CH\(_2\)O PLIF signals than DME hot flames\(^{33}\), as the lack of consumption pathways makes CH\(_2\)O one of the main low-temperature oxidation products.

Figure 5 shows the DME turbulent cool diffusion flame structure for a main flow velocity of \( U = 1.50 \) m/s and a DME mole fraction of \( X_{\text{DME}} = 0.2 \). This condition corresponds to a turbulence intensity of \( u' / U = 0.16 \). All three streams (main, pilot, co-flow) exit the burner at a temperature of \( T = 600 \) K (as elucidated in Fig. 4). The turbulent cool flame is clearly wrinkled but unbroken (Fig. 5a), with the majority of the wrinkling occurring on the fuel side of the flame. The higher likelihood of fuel-side fluctuations is confirmed quantitatively by the values of the standard deviation of the CH\(_2\)O signal in Fig. 5c.

When the main flow velocity is increased to \( U = 2.75 \) m/s, pockets of unburned reactants and flame islands become much more common (Fig. 6a). The strength of the average CH\(_2\)O signal (Fig. 6b) decreases noticeably compared to the \( U = 1.50 \) m/s case, especially in the regions closest to the burner exit. Fig. 6c also reveals that the standard deviation of the CH\(_2\)O signal generally decreases on the fuel side of the flame. However, fluctuations are seen to substantially increase on the oxidizer side, particularly for the regions furthest from the burner exit. Thus, at

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increased levels of turbulence, two distinct maxima in the fluctuating values occur, which resembles measurements of DME turbulent hot diffusion flames\textsuperscript{24}.

\section{Flame Structure Measurements through Rayleigh Scattering}

Figure 7 shows the DME turbulent cool diffusion flame temperature distribution at $x/R = 2.2$ for a main flow velocity of $U = 1.50$ m/s and $X_{\text{DME}} = 0.2$ (identical conditions to Fig. 5). As alluded to previously, the cool flame temperatures are determined by comparing the raw Rayleigh scattering signal at $T = 600$ K (cool flame) to the raw signals from a $T = 500$ K unburning case and a $T = 550$ K unburning case. Besides the exit temperatures, all three cases have identical boundary conditions ($X_{\text{DME}}, U, U_{\text{pilot}}, \text{etc.}$).

The temperature of the central $\text{N}_2$/DME stream at an axial location of $x/R = 2.2$ is determined to be $T \approx 595$ K, which is quite close to the value of $T = 600$ K measured by thermocouple at $x/R = 0$. At $x/R = 2.2$, the maximum mean temperatures occur near $r/R \approx \pm 0.9$. The maximum temperature is slightly asymmetric, peaking at $T \approx 725$ K on left side and $T \approx 700$ K on right side.

Fig. 5. $\text{CH}_2\text{O}$ PLIF measurements for DME turbulent cool diffusion flames at $U = 1.50$ m/s and $X_{\text{DME}} = 0.2$. Instantaneous values are shown in (a), mean values in (b), and standard deviations in (c).

Fig. 6. $\text{CH}_2\text{O}$ PLIF measurements for DME turbulent cool diffusion flames at $U = 2.75$ m/s and $X_{\text{DME}} = 0.2$. Instantaneous values are shown in (a), mean values in (b), and standard deviations in (c).
Like the CH$_2$O PLIF measurements at the same condition (Fig. 5), the maximum standard deviation in the temperature ($T' \approx 90$ K) occurs closer to fuel side ($r/R \approx \pm 0.8$) than the maximum mean temperature. Interestingly, the maximum temperature fluctuations in Fig. 7 are very close to the values obtained from thermocouple measurements in an $n$-heptane turbulent cool premixed flame. In this study, the minimum temperature fluctuations ($T' \approx 20$ K) occur in the unburning regions away from the flame. Together, these measurements give $T'/T \approx 0.03$ in the unburning regions and up to $T'/T \approx 0.13$ in the cool flame itself.

VI. Conclusion

A new Co-flow Axisymmetric Reactor-Assisted Turbulent (CARAT) burner has been utilized to study turbulent cool diffusion flames. Measurements with a nanoscale thermal anemometry probe (NSTAP) reveal that the burner is capable of producing turbulence intensities from $0.16 < u'/U < 0.20$ over the range of conditions studied. By heating the co-flow (air), pilot (oxygen), and main (nitrogen/dimethyl ether) streams to $T = 600$ K, turbulent cool diffusion flames are able to be established over a range of conditions. Formaldehyde planar laser-induced fluorescence measurements indicate that weak turbulence causes wrinkling to occur primarily on the fuel side of the cool flame. However, increased turbulence is seen to produce significant fluctuations on both sides of the flame. Measurements of the cool flame temperature, obtained through Rayleigh scattering, confirm these structural observations for weak turbulence. The experimental methodology described in this study provides a well-defined platform for the study of turbulence-chemistry interactions at low temperatures.

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