Effect of quenching of the oxidation layer in highly turbulent counterflow premixed flames

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Abstract

Three lean-to-stoichiometric premixed flames were studied in the opposed-jet configuration by counterflowing fresh reactants and fully burnt products of combustion at 1850 K. The study was performed experimentally under turbulent conditions and computationally under laminar ones. The flame mixtures were selected to have the same unstrained laminar flame speed. The turbulent flames were studied experimentally using simultaneous planar imaging of CO and OH laser-induced fluorescence, which, in combination, yielded a quantity proportional to the forward reaction rate for CO + OH $\rightarrow$ CO$_2$ + H. Since the turbulence characteristics of the feed streams were identical for all flames, the turbulent burning regime was also the same, with a turbulent Reynolds number of 1050 and a Karlovitz number of approximately 5. The oxidation layer of the stoichiometric flame was extinguished, whereas the lean flames exhibited substantial evidence of CO conversion as indicated by the CO + OH reaction rate imaging. To aid the interpretation of the experiments, we numerically investigated the extinction of strained laminar premixed flames with compositions identical to those of the experiments. The calculations corroborated the experimental results, indicating that the stoichiometric flame was the least robust and extinguished at the lowest strain rate. Furthermore, extinction occurred when the flames were very close to the gas stagnation plane and the oxidation layer extended beyond it, towards the burnt product side. The quenching of the oxidation layer is suggested as a possible reason for either local or overall extinction of highly strained premixed flames.

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1. Introduction

From a flamelet perspective of turbulent combustion, a theory of turbulent premixed combustion based on the assumption of scale separation typically compares the thicknesses of the preheat zone and the fuel-consumption layer of the laminar premixed flame with relevant turbulence length scales, such as the Kolmogorov microscale [1]. A turbulent Karlovitz number $K_a$, defined as the ratio of a relevant chemical time scale to the turn-over time of the Kolmogorov microscale, is often used to quantify the degree of turbulence/chemistry interaction. It is expressed as a function of the turbulent velocity fluctuations $\nu'$, the laminar
flame speed $S_L$, the integral length scale $l'$ and the flame thickness $l_F$, as:

$$K_a = \frac{t_{ch}}{l_{ch}} = \left( \frac{l'}{S_L} \right)^{3/2} \left( \frac{l_F}{T} \right)^{1/2}$$

in which $l_F$ is typically scaled as $D/S_L$, in the simplified context of one-step chemical kinetics, with $D$ as the thermal diffusivity estimated for instance at the adiabatic-flame temperature. When $K_a$ is $O(100)$, departures from the flamelet regime with local extinction of the flame front are anticipated. Such conditions of strong turbulence and chemistry interaction are particularly challenging to reproduce in a laboratory scale burner. Techniques such as intense swirling or pilot flames have been used to stabilize highly turbulent premixed flames, but evidence of the non-flamelet regime is relatively rare [2,3].

Whereas the ability of turbulent eddies to enhance the transport within the preheat zone and to modify the thickness of the flame thermal layer was previously observed [4,5], the effect of turbulence on the oxidation layer and the role of the CO-to-CO$_2$ conversion rate in turbulent premixed flames have not been investigated experimentally. Direct numerical simulations have demonstrated that CO oxidation is susceptible to unsteady strain rate effects [6]. The reason for the surprisingly modest interest in the dynamics of the oxidation layer may be twofold [1]: first, the feedback of the oxidation layer on the fuel-consumption layer, where most of the heat release is produced, is generally assumed to be weak; second, the turbulent intensity is weakened in the hot oxidation layer because of the increase in the gas kinematic viscosity. As a result, turbulent eddies are not expected to have a significant effect on the structure of the oxidation layer.

The goal of the present contribution is to monitor the oxidation layer in turbulent premixed flames and its possible role in the extinction process. To that end, the counterflow geometry was employed as in previous investigations of turbulent premixed flames [7–9]. A novelty of the present burner lied in the turbulence generation scheme enabling turbulent Reynolds number of $O(1000)$ [10]. Well-controlled heat losses and mean strain rates are imposed on turbulent premixed flames by counterflowing jets of fresh reactants and hot combustion products, a configuration previously used in [11]. The temperature of the products and the separation between the nozzles define the volumetric heat loss and the bulk strain rate, respectively. We selected three lean-to-stoichiometric CH$_4$/O$_2$/N$_2$ premixed flames with identical laminar flame speeds but different structures of the oxidation layer by varying the equivalence ratio and the amount of inert dilution of the three flame reactant mixtures. Remarkably, despite the fact that the three flames have virtually identical $K_a$ and coordinates in a modified Borghi diagram [1], they present different phenomenologies, which points to the inadequacy of this oversimplified framework of turbulent combustion to represent actual turbulent flame dynamics. The article begins with a description of the experimental apparatus. Selected experimental results from the turbulent flames are then presented and are interpreted with the help of laminar flame calculations for the same feed stream composition.
frequencies in the turbulent power spectrum, and preserving the axisymmetric uniformity of the mean flow field [12]. The lower nozzle was specifically designed to produce a jet of combustion products at elevated temperature using a pre-burner consisting of a cylindrical ceramic combustion chamber terminating in a convergent nozzle, in which a flame was ignited 155 mm upstream of the nozzle outlet to ensure completeness of combustion by the time the stream exited the nozzle. The fuel/oxygen ratio of this pre-burner was fixed at unity stoichiometry and the oxidizer mixture contained between 71% and 74% nitrogen with the balance in oxygen, to maintain the temperature of the combustion products at 1850 K. The large viscosity of the hot combustion products resulted in a practically laminar flow issuing from the lower nozzle.

The reactant equivalence ratio \( \varphi \) and nitrogen content of the ‘air’ stream \( X(N_2) \) are specified in Table 1, along with the adiabatic-flame temperature \( T_{ad} \), the laminar flame speed \( S_l \), the thickness of the oxidation layer \( \delta_{oxl} \) (see Section 4.2 below), and other quantities of relevance to the characterization of the turbulence such as: \( \nu / S_l \), \( \delta_{oxl}/\ell \), \( Re_l \) and \( Ka_t \).

### 2.2. CO/OH reaction rate imaging

Simultaneous imaging of single-photon OH laser-induced fluorescence (LIF) and two-photon CO LIF was used to measure the relative forward reaction rate of the CO + OH \( \rightarrow \) CO\(_2 + \) H reaction. This diagnostic approach enables us to probe the CO-to-CO\(_2\) conversion rate in CH\(_4\)/O\(_2\)/N\(_2\) flames. The reaction rate imaging technique is described in detail elsewhere [13–15], and only a brief overview is given here. The forward reaction rate, \( RR \), is given by \( RR = k(T)\text{CO}[OH] \), where \( k(T) \) is the temperature dependent rate constant. The product of the LIF signals from CO and OH can be approximated by \( f_{\text{CO}}(T)f_{\text{OH}}(T)/[\text{CO}[OH]] \), where the temperature dependence of the LIF signals is represented by \( f(T) \). The pump/detection scheme determines the temperature dependence of the LIF signals and can be selected such that \( f_{\text{OH}}(T)/f_{\text{CO}}(T) \propto k(T) \). When this relationship is achieved, the pixel-by-pixel product of the OH LIF and CO LIF signals is proportional to the reaction rate. The accuracy of this correlation for the LIF excitation and detection schemes described below was estimated by calculating variations in quenching rates [16,17] and Boltzmann fraction populations using the unstrained laminar flame calculations in Section 3.2. Over the temperature range of significant CO + OH reaction (1600–2400 K), the proportionality of \( f_{\text{OH}}(T)/f_{\text{CO}}(T) \) to \( k(T) \) is excellent for the three flames considered here with an estimated deviation on the order of ±5%, which is within the uncertainty of the estimate.

For OH LIF measurements, the frequency-doubled output from a 10 Hz Nd:YAG-pumped dye laser was tuned to 285.88 nm to pump the \( Q_1(13) \) transition of the A–X(1, 0) band of OH. An intensified CCD camera (512 \( \times \) 512 pixels) with an f/1.8 Cerco quartz camera lens was used to record the OH LIF signal. The image intensifier was gated for 500 ns bracketing the dye laser pulse. The OH LIF images were corrected for the average spatial variation in the laser sheet using acetone LIF to record the beam profile. The combination of a UG11 color glass filter and a high-transmission bandpass filter (\( T > 80\% \), \( \lambda = 305–325 \) nm) transmitted the OH fluorescence and blocked out-of-band interferences.

Two-photon CO LIF was excited by pumping overlapped transitions in the \( B^1\Sigma_u^- \leftrightarrow X^1\Sigma_u^+ \) (\( \nu' = 0, \nu'' = 0 \)) Hopfield–Birge system of CO using the frequency-doubled outputs from two separate 10 Hz Nd:YAG-pumped optical parametric oscillators (OPO) (14 mJ and 16 mJ/pulse at \( \lambda = 230.1 \) nm). A cylindrical lens was used to combine the laser beams into a 10-mm high laser sheet. The CO fluorescence was imaged onto an intensified CCD camera (512 \( \times \) 512 pixels) using a combination of a medium-format camera lens (f.l. = 80 mm, f/2) and a 35-mm format lens (f.l. = 58 mm, f/1.2). The imaging system included a dual-band interference filter (\( \Delta \lambda = 10 \) nm per band) that blocked out-of-band interference and transmitted fluorescence from the \( B^1\Sigma_u^- \rightarrow \text{A}^1\Pi(\nu' = 0, \nu'' = 1, 2) \) transitions of CO at \( \lambda = 483.5 \) nm and 519.8 nm. For the lean and stoichiometric flames considered here, the dual-band filter provided greater signal collection than a single-band filter with negligible interference from laser-generated C\(_2\) Swan band emission. The image intensifier was gated for 500 ns, bracketing the OPO laser pulses. Shot-to-shot variations in the beam profile were recorded by splitting off a portion of the 230-nm beams after the cylindrical lens and focusing them into a cell containing CO and He at atmospheric pressure and ambient temperature. The CO LIF signal from the cell was recorded on an unintensified CCD camera, and

<table>
<thead>
<tr>
<th>Flames</th>
<th>( \varphi )</th>
<th>( X(N_2) ) in ‘air’</th>
<th>( T_{ad} ) (K)</th>
<th>( S_l ) (cm/s)</th>
<th>( \delta_{oxl} ) (mm)</th>
<th>( \nu / S_l )</th>
<th>( \delta_{oxl}/\ell )</th>
<th>( Re_l )</th>
<th>( Ka_t )</th>
</tr>
</thead>
<tbody>
<tr>
<td>F1.00</td>
<td>1.000</td>
<td>0.75</td>
<td>2392</td>
<td>54.5</td>
<td>5.4</td>
<td>7.25</td>
<td>1.30</td>
<td>1.050</td>
<td>5.0</td>
</tr>
<tr>
<td>F0.70</td>
<td>0.700</td>
<td>0.70</td>
<td>2255</td>
<td>52.9</td>
<td>3.2</td>
<td>7.50</td>
<td>0.76</td>
<td>1.050</td>
<td>5.1</td>
</tr>
<tr>
<td>F0.58</td>
<td>0.575</td>
<td>0.65</td>
<td>2203</td>
<td>51.9</td>
<td>2.4</td>
<td>7.60</td>
<td>0.57</td>
<td>1.050</td>
<td>5.2</td>
</tr>
</tbody>
</table>
the resulting images were used to correct the CO LIF measurements in the counterflow flames for beam profile variations. Additional uncorrected variations in the beam profile arise from beam steering as a result of the laser beam traversing the index of refraction gradients in the turbulent flame.

Timing of the laser pulses was controlled with digital delay generators. The OPO lasers fired 200 ns apart, and the dye laser fired 700 ns before the first OPO to eliminate the possibility of cross-talk between the two diagnostic systems. The CO/OH LIF measurements were essentially instantaneous because the elapsed time for a single measurement was significantly less than the prevailing flow timescale.

The reaction rate imaging technique requires careful matching of the OH and CO LIF images. A precise image matching technique was used to obtain accurate registration between the two CCD cameras [13]. Images were matched with a bilinear geometric warping algorithm, and the residual matching error was in the subpixel range.

For each flame, a series of 300 simultaneous OH and CO LIF images were recorded. An algorithm was designed to compute conditional statistics along the turbulent flame front. The raw images were corrected for background and spatial inhomogeneities of the laser beam. The OH LIF measurements were also corrected for laser light absorption and were smoothed to reduce noise using a single-pass Wiener filter [18] that adapted to the local mean and standard deviation of $5 \times 5$ px$^2$ regions.

3. Results and discussion

3.1. Turbulent premixed flames

The turbulence characteristics of the flow field were measured using particle imaging velocimetry (PIV) in a non-reactive counterflow of hot combustion products at 1850 K and nitrogen at 295 K. Figure 2a shows the mean and RMS axial velocities as a function of the axial coordinate and the axial turbulence intensity as a function of the radial coordinate, at 0.5 mm downstream of the top burner. Under the present conditions, the mean bulk strain rate as estimated from the mean axial velocity at the burner outlet and the burner separation was about 1550 s$^{-1}$. Velocity fluctuations ranging from 3.9 to 9.2 m/s were measured along the burner centerline, and an axial turbulence intensity of approximately 30% was measured near the cold burner mouth and was uniform in the radial direction.

The CO and OH LIF measurements were applied to the three turbulent premixed flames with mixture compositions listed in Table 1. The flames were operated under the same flow conditions with a turbulent Reynolds number of 1050 that was estimated at the exit of the fresh reactant nozzle and a turbulent Karlovitz number of nearly 5, which, in principle, would correspond to the onset of the thin reaction zone regime [1]. Examples of simultaneous two-dimensional LIF images of CO and OH and of the CO + OH reaction rate (RR) are shown in Fig. 3. One example is presented for each flame to illustrate its general topology. The field of view of the CO LIF measurements in flame F1.00 was narrower in the vertical direction as compared to the other two flames because of a problem with one of the lasers for this data set. The lower half of the CO and OH LIF images contains a region of weak signal that corresponds to the counterflowing products of combustion where CO and OH are expected to be in thermodynamic equilibrium. With the fresh reactant mixture being fed from above, the turbulent flames were always located at the upper boundary of the counterflowing products and were easily detected by larger CO and OH LIF signals. The signal contrast between flame products, reactants, and hot counterflowing products was used to identify the position of the mixing layer where the two opposed jets intercepted. In flame F1.00, the production of super-equilibrium OH radical concentrations was not detected, and CO appeared in modest concentrations near the opposed-jet mixing layer, which suggests that the
oxidation layer was completely extinguished. In contrast, flames F0.58 and F0.70 are qualitatively different from the stoichiometric flame. For these lean flames, the CO + OH reaction rate was active and there was evidence of local extinction in the oxidation layer as manifested by discontinuities in the reaction rate contour.

To evaluate these observations on a statistical basis, we compare conditionally averaged CO LIF, OH LIF, and RR profiles of the turbulent flame front for all three flames. To calculate these quantities, the flame front contour was detected in each single-shot measurement by thresholding the OH LIF images. The threshold value was less than the OH LIF signal in the counterflowing products of combustion. Therefore, locally extinguished regions of the flame front were included in the conditional statistics. Every 0.5 mm along a flame front contour, the CO LIF, OH LIF, and RR profiles were sampled along the direction of the local OH gradient, i.e. in the direction normal to the flame front. Each measured profile was interpolated between pixels to facilitate the averaging of profiles conditioned on the local flame-normal coordinate.

Figure 4 shows conditionally averaged flame-normal profiles that were calculated from 300 single-shot measurements. The average CO LIF, OH LIF, and RR curves were normalized by their respective peak values for flame F0.58. The conditionally averaged structures of flames F0.58 and F0.70 were similar to that of a laminar premixed flame. They are indicative of a flame propagating towards the stream of fresh reactants, which is not a foregone conclusion for premixed flames under high strain rate. The conditionally averaged OH LIF profile exhibits a steep gradient where the CO LIF profile reaches a maximum. As a result, the CO + OH reaction rate increases rapidly before decaying slowly downstream, which confirms the presence of oxidation layers in both F0.58 and F0.70. In addition, freely-propagating laminar flame calculations indicate that the CO + OH reaction rate profiles of the three flames considered here are identical. Yet, the conditionally averaged CO + OH reaction rate measurements in flame F0.58 is larger than in flame F0.70, which implies that the chemically active oxidation layers of the latter are more frequently extinguished. Conversely, the oxidation layer is altogether absent in the conditionally averaged structure of flame F1.00. The smooth OH LIF profile is the result of diffusion of the OH radical in the counterflowing products of combustion across the turbulent mixing layer. Therefore, the primary path of conversion of CO into CO2 is virtually non-existent in such a flame. In freely propagating flames, this lack of conversion would correspond to the loss of one-third of the total heat release. As shown in the following subsection, a flame for which the oxidation layer is quenched is at the very onset of extinction.

3.2. Extinction of strained laminar premixed flames

To guide the interpretation of the experimental results within the flamelet context, the extinction of the three premixed flames was also investigated computationally using a detailed chemistry mechanism and transport model. The simulated problem was the steady-state laminar counterpart of the experimental system and consisted of counterflowing laminar jets of fresh reactants and hot products of combustion at 1850 K. The elliptic form of the 2D conservation equations in cylindrical
coordinates was reduced to a 1D nonlinear two-point boundary value problem by making use of a similarity transformation. The reduced pressure gradient $J = (1/r)(\partial P/\partial r)$, with $P$, the pressure, and $r$, the radial spatial coordinate, was solved as an eigenvalue of the system. A phase-space, pseudo-arclength continuation method was employed, where the two inlet axial momenta were used as continuation variables. The strain rate $SR$ was derived from $SR = (J/\rho_u)^{0.5}$, with $\rho_u$, the density of the fresh reactants. Details of the model are provided in [19–21]. Selected results of these numerical simulations are presented. Figures 5a and b show the heat release rate per unit flame area and the position of the peak heat release rate as a function of strain rate for three laminar flames with compositions identical to those of the turbulent flames in Section 3.1. Extinction was determined at the turning point, where the slope of the flame heat release rate is vertical. Even though the unstrained laminar flame F1.00 releases more heat than the other flames, it loses heat faster with increasing strain rate and extinguishes at a significantly smaller strain rate (Fig. 5a). An increase in strain rate shifts the flame towards the gas stagnation plane, and extinction occurs when the location of peak heat release is less than half a millimeter from the stagnation plane, but still on the fresh reactant side of such a plane (Fig. 5b). At low strain rates, the three flames are located at the same position since they have essentially the same unstrained laminar flame speed. Near extinction, however, the flames have different positions.

Figure 6 shows that, as a premixed flame approaches the gas stagnation plane, the peak heat release rate is at first modestly reduced, but, once the flame is within a couple of millimeters from the stagnation plane, the heat release drops significantly. A recent study [22] demonstrated that major species, of the stoichiometric counter-flowing products, such as $\text{N}_2$, $\text{CO}_2$ and $\text{H}_2\text{O}$, dilute the flame once it reaches the vicinity of the stagnation plane. This effect combined with heat losses induced by the sub-adiabatic temperature of the hot combustion products affect the oxidation layer well before the fuel-consumption layer, as illustrated in Fig. 6 for flame F1.00 with a series of heat release profiles up to the onset of extinction at 905 s$^{-1}$. The fuel-consumption layer coincides with the peak heat release rate and the oxidation layer corresponds to the long tail that extends towards the stagnation plane. Envelope curves of the loci of peaks of heat release of the three flames are also plotted as a function of strain rate in Fig. 6. They clearly show that flame F1.00 is quenched first as the flames approach the stagnation plane. These results are consistent with the findings of Sanchez et al. [23] who reported that, for lean premixed flames at 18 atm, increases in strain rate reduce the rate of CO-to-CO$_2$ conversion before affecting the rate of fuel consumption. This result was also confirmed in [21] where the cause of extinction of atmospheric premixed flames in the presence of stoichiometric counter-flowing hot products was attributed to the quenching of the oxidation layer with the entire H–O–OH radical pool being reduced.
In Fig. 7, a combustion progress variable, defined as the normalized CO₂ mole fraction \( X_{\text{CO}_2} / X_{\text{CO}_2,0} \), where \( X_{\text{CO}_2,0} \) is the CO₂ mole fraction in the combustion products, is used to compare the thicknesses of the oxidation layers of the three freely-propagating premixed flames. The progress variable \( 1 - X_{\text{CH}_4} / X_{\text{CH}_4,0} \), based on the mole fraction of methane normalized by the value in the fresh mixture, is shown in the same figure for flame F1.00. The curves of the CH₄ progress variable for the two lean flames are not shown but are virtually indistinguishable from that of the stoichiometric flame on the scale of Fig. 7. Since CH₄ diffuses into the preheat zone and is fully consumed in the fuel-consumption layer, the virtual boundary between the fuel-consumption layer and oxidation layer was defined as the location where 99% of the fuel was consumed \( (1 - X_{\text{CH}_4} / X_{\text{CH}_4,0} = 0.99) \) and was chosen as origin of the abscissa in Fig. 7. For reference with the previous figures, the positions of maximum heat release rate of the three flames are identical and located 0.07 mm upstream of the inner-layer boundary. In the fuel-consumption layer of all three flames, the CO₂ concentration reaches nearly 50% of its value in the product region. However, the CO₂ progress variables of flames F0.58 and F0.70 reach unity in a significantly shorter distance as compared to the stoichiometric flame F1.00. For flame F1.00, the thickness of the oxidation layer \( (\delta_{\text{oxl}} = 5.4 \text{ mm}) \), defined as the location where the CO₂ progress variable reaches 99%, is 1.7 and 2.3 times larger than that of flames F0.70 \( (\delta_{\text{oxl}} = 3.2 \text{ mm}) \) and F0.58 \( (\delta_{\text{oxl}} = 2.4 \text{ mm}) \), respectively, as reported in Table 1. In comparison, the thickness of the fuel-consumption layer was 0.33 mm for all flames. Consequently, at low strain rate where the peak of heat release of the three flames occupies the same position, the oxidation layer of F1.00 extends closer to the stagnation plane than the oxidation layers of the lean flames, and F1.00 loses heat faster than the other flames.

It is tempting to identify a modified turbulent Karlovitz number, \( K_{\text{oxl}} \), in which the thickness of the oxidation layer, as estimated from laminar flame calculation, is used in Eq. (1), instead of the flame thickness, which would result in the stoichiometric flame having indeed the largest Karlovitz number \( K_{\text{oxl}} \). However, further experimental corroboration is necessary before attempting to capture the complexities of heat loss/gain and flame dilution by the counterflowing hot products using a single non-dimensional number.

4. Conclusions

An experimental study was conducted on highly turbulent flames established by counterflowing unreacted premixtures with hot combustion products. Turbulence conditions were identical in all flames, with \( O(1000) \) turbulent Reynolds number, and the mixture compositions were selected to ensure that the laminar flame speed was constant, even though the flames had equivalence ratios ranging from lean to stoichiometric. As a result, all flames occupied the same position in the extended Borghi diagram. Yet, they exhibited qualitative differences, as evidenced by OH and CO LIF imaging measurements that provided a measure of the forward reaction rate of \( \text{CO} + \text{OH} \rightarrow \text{CO}_2 + \text{H} \). The oxidation layer of the stoichiometric flame was quenched, unlike the lean flames. Furthermore, the absence of measurable OH LIF signal in the stoichiometric “flame” suggested that the entire “flame” was quenched, with only modest heat release that was associated with residual fuel oxidation to form measurable amounts of CO. This interpretation was substantiated by calculations performed on strained laminar flames with the same composition, showing that the stoichiometric flame was less robust than the others as it reaches extinction at lower strain rates. These laminar flames extinguished when the flame was within a fraction of a millimeter of the gas stagnation plane, with heat from the oxidation layer diffusing into the burnt product side of the stagnation plane. The study demonstrated the ability of this highly turbulent counterflow system to mimic conditions of relevance to practical combustion and to produce finite-rate chemistry effects that the existing, oversimplified turbulent premixed combustion framework fails to capture.

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