Laminar Flame Speed Measurements of Spherically Expanding CH$_4$-Air Flames from Chemiluminescence of OH* and CH*

Mattias A. Turner, Tyler Paschal, Pradeep Parajuli, Waruna D. Kuletilaka, and Eric L. Petersen
Texas A&M University
College Station, Texas, 77843, USA

1 Introduction

Laminar flame speed is a well-studied fundamental property of a fuel-oxidizer mixture that is commonly used as a metric for validation of chemical kinetics models. Laminar flame speed has been measured by various methods, including using the spherically expanding flame bomb [1–11]. Spherically expanding flame rigs are commonly coupled with schlieren imaging as the primary optical diagnostic method. The density gradient between the high-temperature product gases and the low-temperature reactant gases causes a change in the index of refraction of the gas mixture, creating a region of diffraction that can be recorded using high-speed photography. This region of large density gradient is assumed to be a close approximation of the reaction zone. However, because the schlieren effect is indicative of the density gradient created by combustion and not necessarily of the flame reaction zone itself, it may be more accurate to investigate the reaction zone directly using species-specific chemiluminescence (CL) diagnostics. The electronically exited intermediate species that exist temporarily in the reaction zone of the flame emit unique spectra that can be recorded using a high-speed camera/image intensifier assembly equipped with optical filters to isolate a desired narrow bandwidth of emission. Chemiluminescence imaging is more commonly used for other flame measurement methods, such as stagnation methods, the Bunsen burner method, or the heat flux method, but has not yet been applied to spherically expanding flame vessels to the authors’ knowledge. Dandy and Vosen [12] conducted measurements of OH* chemiluminescence in roughly spherically expanding methane-air flames, but used a photomultiplier tube to capture intensity readings. This method simply produced chemiluminescence intensity per unit projected area.

For the present study, chemiluminescence of OH* and CH* at 306 nm and 430 nm, respectively, was recorded using a Photron Fastcam SA-Z high-speed camera and LaVision High-Speed Intensified Relay Optics (HS-IRO) intensifier. At each equivalence ratio, total broadband emission was also recorded for comparison. Methane was chosen as the fuel for this study due to the large body of literature available with which to compare the new data. The laminar flame speed of methane has been measured in a
spherically expanding flame bomb a number of times using schlieren imaging [1, 2, 4–11]. The focus of this paper was the extraction of laminar flame speed measurements from chemiluminescence images in place of schlieren images. The established image processing analysis of schlieren images can be easily adapted to the analysis of chemiluminescence images, due to the leading edge of the flame being clearly defined in chemiluminescence photography, just as it is in schlieren photography.

2 Experimental Facility and Procedure

The experimental apparatus employed for this work is a fan-stirred flame vessel, designed for initial conditions of up to 10 atmospheres and 400 K. The apparatus is described in depth by Morones et al. [13, 14]. The vessel is a stainless steel, constant-volume vessel with four, 127-mm diameter optical ports, installed with 88.9-mm-thick, fused silica windows to provide two perpendicular axes of optical access. Reactant mixtures were ignited using an automotive-grade ignition coil, with the spark gap located at the center of the volume of the vessel. The spark is created by supplying the ignition coil with a 12-volt power circuit and a normally open, 5-volt triggering circuit. When the 5-volt circuit is closed by the press of a button, the coil discharges and the spark is generated.

As seen in Fig. 1, the optical setup was comprised of a Photron SA-Z high-speed camera coupled to a LaVision HS-IRO unit, and is described in more detail by Paschal et al. [15]. To isolate the desired wavelength for each of the radicals, narrow bandpass optical filters were installed in front of the intensifier. The filter used for OH* was 315 nm ± 15 nm, and the CH* filter was 434 nm ± 17 nm. Broadband emission was also recorded for each equivalence ratio.

Fig. 1  Schematic of full experimental rig, with 5-volt coil trigger routed to delay generator input.

Due to the weak emission of the OH* and CH* radicals, the light must be amplified by the intensifier to raise the intensity of the captured emission. The range of gain values used for this work ranged from 40% to 63% on the internal scale set by the intensifier software. Because the gain was changed for each test, inferences cannot be made regarding the relative quantity of CH* or OH* generated in the flame.

The intensifier is ultra-sensitive to light exposed and must be protected from high-intensity light, including the emission from the spark at the very beginning of the test and emission from the flame near the end of
the experiment. Repeated overexposure of the IRO can cause damage, so a triggering system was designed to prevent the exposure of the IRO to the spark emission. The triggering system used the 5-volt ignition coil trigger signal as its time-zero master trigger. By this method, it was determined that the spark emission remains significantly intense for approximately 13.35 ms, which was then used as the time at which the intensifier shutter was first triggered to open for each experiment. The experimental apparatus, including the triggering system, is depicted in Fig. 1.

Gas mixtures were prepared using the partial pressure method, by which each constituent gas in the reactant mixture was added to the vessel in ascending order of partial pressure. All gases used in this study were ultra-high-purity grade and sourced from Praxair. Before filling the vessel with the reactant mixture, the vessel was evacuated using a roughing pump to a pressure below 0.3 torr. This residual gas is assumed to be air. The vessel was then filled with each gas in ascending order of partial pressure. During the filling process, the temperature of the gases in the vessel was monitored using a thermocouple. The temperature of the gas mixture was allowed to return to within 0.2 K of the initial temperature of the mixture before the next component was added.

3 Data Analysis

The extraction of laminar flame speed from the raw images begins with a conversion of the images to greyscale, if necessary. The images are then processed using an edge-detection algorithm developed in-house, as described by Sikes et al. [4]. A circle is fit to the leading edge of the flame in every image, providing the radius of the flame at known time intervals based on the frame rate of the camera.

Spherically expanding flames are inherently stretched, and therefore the burned, unstretched state of the flame (referred to as $S_b^0$) must be extrapolated from the burned, stretched state captured in the images of the flame (referred to as $S_b$). This process, described by Sikes et al. [4], is completed by plotting the time-rate-of-change of the flame radius, referred to as $dr/dt$, as a function of the stretch rate of the flame. The early growth of the flame is affected by the energy added to the system by the spark, and the late growth of the flame can be affected by confinement of the vessel wall. Therefore, some of the $dr/dt$ values on either end of each data set are excluded from the calculation of laminar flame speed based on the residual of each value of $dr/dt$.

To extrapolate the burned flame speed to a condition of zero stretch, one of two non-linear methods is used, depending on the Markstein length ($L_{m,b}$). The first non-linear method, known as NM I, is used when $L_{m,b}$ is positive, whereas NM II is used when $L_{m,b}$ is negative [4]. Non-linear methods NM I and NM II are included as Equations 1 and 2, respectively. After extracting burned, stretched flame speed from the data, the ratio of the densities of the burned gases ($\rho_b$) and the unburned gases ($\rho_u$) is used to convert $S_b^0$ to the unburned, unstretched flame speed, referred to as $S_L^0$. The densities of the burned gases and unburned gases are calculated using Chemkin’s equilibrium chemistry solver. In Eqs. 1 and 2, $L_{m,b}$ refers to the burned Markstein length, and $r$ refers to the radius of the flame. The Markstein length is determined by the best fit of the appropriate non-linear method to the data.

$$S_b = S_b^0 - S_b^0 L_{m,b} \cdot \frac{2}{r}$$ (1)

$$\ln(S_b) = \ln(S_b^0) - S_b^0 L_{m,b} \cdot \frac{2}{rS_b}$$ (2)

$$S_L^0 = S_b^0 \frac{\rho_b}{\rho_u}$$ (3)
Results and Discussion

Figure 2 presents the new laminar flame speed data for methane at 1 atm and room temperature (21°C, 294 K) for each of the three emission markers – CH*, OH*, and broad band. The estimated uncertainty in the data in this work is ±5%. Each of the three wavelength bandwidths produced similar flame speeds at each equivalence ratio. Therefore, the flame speeds derived from the chemiluminescence data at each equivalence ratio was averaged to simplify the comparison with the literature data. As shown in Fig. 2, the new chemiluminescence method produces laminar flame speeds that agree well with established literature for every equivalence ratio tested. The literature data reproduced in Fig. 2 were taken from Aung et al. [5], Bosschart and de Goey [3], Gu et al. [2], Hassan et al. [1], Lowry et al. [6], Rozenchan et al. [10], Sikes et al. [4], Tahtouh et al. [9], and Vagelopoulos and Egolfopoulos [8]. For clarity and legibility, all of the literature data reproduced herein were measured using a spherically expanding flame apparatus, with the exception of the data reported by Bosschaart and de Goey (heat flux) and Vagelopoulos and Egolfopoulos (flat flame).

The images for each targeted wavelength of chemiluminescence were compared for each equivalence ratio to determine qualitatively the differences between the types of chemiluminescence. An example of this comparison is shown in Fig. 3, where each type of chemiluminescence is compared for three timestamps at the stoichiometric equivalence ratio. There are several differences between the types of chemiluminescence that are worth noting. First, the distribution of intensity across the flame is unique for each wavelength. The broadband emission is very bright at the edge of the flame and only fades slightly at center of the flame, whereas the CH* chemiluminescence is much less intense at the center of the flame. The OH* emission is somewhat brighter at the leading edge of the flame than it is toward the center of the flame, and it increases in intensity in the center of the flame as the flame grows. This gradient indicates that OH* is present in the interior of the flame even after the initial flame front passes. The OH* images also exhibit artifacts of the spark for much longer than the other wavelengths. The CH* images are notable.

Fig. 2 Left: Laminar flame speed of methane-air measured from different wavelengths of chemiluminescence at one atmosphere and room temperature for a range of equivalence ratios. Right: Laminar flame speed of methane-air from this study plotted with literature data for one atmosphere and room temperature for a range of equivalence ratios.
for exhibiting a clearly defined, bright leading flame edge that lends itself well to the application of the same routines used for the previously existing schlieren image processing. A higher gain was required for the CH* images as compared to the OH* and broadband images. The broadband images tended to be more saturated than the OH* or CH* images, even with the low-gain settings. This bright intensity caused a glow to appear around the flame in many of the late-flame images, as can be seen in the 30.35-ms image of broadband chemiluminescence in Fig. 3. It is also worth noting that these chemiluminescence images capture many of the physical features of the flame that make the schlieren measurement of spherical flames visually interesting.

Fig. 3 Images from three tests conducted at the stoichiometric equivalence ratio for the three different emission bandwidths. The time stamps correspond to each column, and the emission wavelength labels correspond to each row.

Because the broadband emission cannot be traced back to any single species, its utility is limited simply to laminar flame speed measurements and qualitative image analysis. For this reason, it will be more practical to select a single wavelength at which to continue this testing that can be directly related to the concentration or population of a radical of interest, so that this type of imaging can be used for additional analysis, such as flame reaction zone thickness [15]. OH* and CH* chemiluminescence both show promise in this regard. CH* may have an advantage over OH* because of the higher contrast between the leading edge of the flame and the center of the flame and because of the lack of residual spark emission in the CH* images.

5 Conclusion

Chemiluminescence imaging can produce detailed images of spherical flames, from which laminar flame speed and chemical species time histories can be extracted, but additional work is needed to fully validate the technique. Laminar flame speeds of methane-air flames measured from chemiluminescence agree well with the established spherical-flame literature data, which has been traditionally recorded by schlieren photography. In the future, both optical axes of the authors’ experimental apparatus will be utilized simultaneously to conduct time-synchronized schlieren photography and chemiluminescence photography of a single combustion event. This configuration will provide a set of data in which the differences between the two imaging techniques can be compared regarding the measurement of laminar flame speed.
References