Joint Quantitative Imaging of Rayleigh and OH LIF Using Saturated and Linear Fluorescence

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ABSTRACT

An experimental technique is described that enables quantitative evaluation of simultaneous 2-D temperature and OH radical distributions. The aim of this technique is to provide a quantitative investigation technique to be applied to the Piloted Premixed Vitiated Coflow Burner (PPVCB) described by Dunn et al. [4].

Significant effort has been made to eliminate errors that commonly plague the quantification of the OH Planar Laser Induced Fluorescence (PLIF) and Rayleigh signal. For the Rayleigh signal, factors such as laser power fluctuations, background reflections and non uniform cross section have been accounted for. A new model is presented allowing quantification of the OH mole fraction in both the linear, transition and saturated regimes of fluorescence. Demonstration of the experimental technique as a proof of principle in the PPVCB is presented.

1. INTRODUCTION

Calculating turbulent premixed flames remains a challenge due to the difficulty of representing the burning rates and the lack of a universal model relevant over the entire regime diagram. Although there are different interpretations of the regime diagrams e.g. Chen et al. [2] and Peters [13], most of them attempt to correlate in non-dimensional form the flame structure (thickness and burning velocity) to the turbulence characteristics (intensity and length scale). Given these limitations it is important to understand local flame structure and hence its location on the regime diagram, as this influences the choice of combustion model.

Typically for premixed combustion, experimental data consists of simultaneous OH PLIF and 2-D Rayleigh; one example of this method is reported by Chen et al. [1]. This enables a measure of a reaction front marked by either temperature and/or OH. A reaction progress variable may also be derived based on temperature and OH concentration. Using this experimental method a number of issues addressed (Chen et al. [2]), such as:

- In the wrinkled flamelet regime does the OH concentration vs. temperature show a similar structure to that in strained laminar flames?
- In the turbulent flame-front regime as defined by Chen et al. [2], under what conditions are the isolated island patches of C<sub>t</sub>=0.5 burning or extinguishing?
- In the stretched flame front regime what is the shape and thickness of the flame front with respect to flow structures?

Using the PPVCB burner described by Dunn et al. [4] and a sound experimental method it is hoped that we will be able to further validate and refine the understanding of the turbulent premixed combustion regime diagram. Addressing the need for the experimental data being quantitatively accurate is the particular interest of this paper, with a proof of principle applied to a couple of representative flames. Particular effort has been made to make the OH fluorescence signal quantitative, utilising both the saturated and linear regimes of the beam profile.

2. Experimental Setup

The experimental setup consisted primarily of forming an overlapping Rayleigh and UV 2-D sheet at the probe volume, and collecting and filtering the appropriate signal. The Rayleigh laser sheet was generated from the second harmonic of a Spectra Physics Pro350 Nd:YAG, operating at an energy of 130mJ/pulse. The 283.2 nm UV beam at 28mJ/Pulse, corresponding to Q<sub>4</sub>(7) excitation for the A<sup>2</sup>S<sup>-</sup>-X<sup>2</sup>Π (1,0) system of OH-LIF, was generated from the second harmonic of a seeded Spectra Physics Pro350 Nd:YAG pumping the frequency doubled fundamental of a Syrah Pumped Dye Laser (PDL) using Rhodanine 6G in ethanol.

![Diagram](image.png)

Figure 1. The laser excitation and detector setup, used for the experiment.

Of critical importance when measuring scalar gradients in turbulent flows is to obtain a laser sheet that has a minimal focal waist thickness, to ensure that spatial gradients are adequately resolved. The laser beam thickness at the focus and edge of the image was estimated by using the laser burn marks produced on photosensitive paper. Considering a Gaussian intensity field laser beam, inherent laser beam divergence, laser beam diameter and optics used, the calculated diffraction limit for the focal point is 19µm and 30µm for the 283nm and 532nm beams respectively. Measuring under a microscope the laser beam burn marks at the focal point were found to be 20µm (± 7µm) and 40µm (± 8µm) for the 283nm and 532nm beams respectively. The burn mark measurements, particularly the 283nm marks are very close to the calculated diffraction limit, the authors acknowledge the limitations of obtaining...
absolute measurement accuracy using the burn mark method. The burn mark method does however give a definite indication of the focal waist thickness.

3. Image processing

3.1 Rayleigh image processing

The magnitude of the Rayleigh scattering signal received by pixel \(m, n\) on the detector ICCD from the respective probe volume may be expressed as:

\[
R = n L_{\text{Laser}} \Omega N \sum_{i=1}^{N} X_i \sigma_i
\]

(1)

Where \(n\) is the collection efficiency of the detection system that includes quantum efficiency of the detector, \(L_{\text{Laser}}\) is the laser intensity, \(\Omega\) is the solid collection angle of the detection system, \(N\) is the total molecular number density, \(X_i\) is the mole fraction of species \(i\) and \(\sigma_i\) is the cross section for species \(i\).

In order to process raw ICCD Rayleigh images and derive quantitative images for temperature, corrections need to be made for: spatial non-uniformity in laser irradiance, non uniform cross section, detection system dark noise, shot to shot fluctuations in laser power and background reflections. As a start to implementing the said corrections, the raw Rayleigh image \(I_{\text{RawRayleigh}}\), may be expressed as:

\[
I_{\text{RawRayleigh}} = I_{\text{Background}} + I_{\text{Reflection}} + I_{\text{Rayleigh}}
\]

(2)

Where both the image components \(I_{\text{Reflection}}\) and \(I_{\text{Rayleigh}}\) are both linearly proportional to the individual shot’s laser power. The image component \(I_{\text{Background}}\) which represents the background due to detection system dark-noise is easily evaluated for identical ICCD gate timing and gain settings without the laser pulse. For an ensemble average of images with a known, fixed cross section the ensemble average will be representative of an average laser power. Utilising the relation developed in (2) and two ensemble average Rayleigh images taken in a known medium of air or helium, the following Equations (3-4) may be developed.

\[
I_{\text{Air}} = I_{\text{Background}} + L_{\text{Avg}} K_{\text{Reflection}} + C_0 N_{\text{Air}} L_{\text{Avg}} \sigma_{\text{Air}}
\]

(3)

\[
I_{\text{He}} = I_{\text{Background}} + L_{\text{Avg}} K_{\text{Reflection}} + C_0 N_{\text{He}} L_{\text{Avg}} \sigma_{\text{He}}
\]

(4)

The calibration constant \(C_0\) is determined experimentally. Equations (3-4) may be rearranged to obtain an equation for the extrapolated zero Rayleigh cross section signal, which represents the laser reflection constant \(K_{\text{Reflection}}\) for an average laser power.

\[
K_{\text{Reflection}} = \frac{(I_{\text{He}} - I_{\text{Air}} \sigma_R)}{(1 - \sigma_R)} - I_{\text{Background}}
\]

(5)

\[
\sigma_R = \frac{\sigma_{\text{He}}}{\sigma_{\text{Air}}}
\]

(6)

Zhao et al [16] in a review of Rayleigh scattering measurements discusses the evaluation of the background reflections however doesn’t extend this concept to include fluctuations in the background reflections with laser power as proposed in Equations (3-4).

In order to compensate for fluctuations in \(I_{\text{Reflection}}\) and \(I_{\text{Rayleigh}}\) due to laser power fluctuations in the flame shot \(I_{\text{FlameShot}}\), the shot laser power \(L_{\text{Shot}}\) needs to be evaluated. This is done by taking the average of a small area in each image near the laser beam centreline, and comparing the same area in the ensemble average of flame image. Close to the edge in the image the coflow composition can be considered to be uniform and not to fluctuate or be influenced by the central jet combustion process. The values in the area of the coflow in the individual and ensemble average images can be expressed as:

\[
I_{\text{FlameAvg}} = I_{\text{Background}} + L_{\text{Avg}} K_{\text{Reflection}} + C_0 L_{\text{Avg}} \sigma_{\text{Coflow}}
\]

(7)

\[
I_{\text{FlameShot}} = I_{\text{Background}} + L_{\text{Shot}} K_{\text{Reflection}} + C_0 L_{\text{Shot}} \sigma_{\text{Coflow}}
\]

(8)

The individual Rayleigh shot image laser power may be evaluated as:

\[
L_{\text{Shot}} = \frac{I_{\text{FlameShot}} - I_{\text{Background}}}{I_{\text{FlameAvg}} - I_{\text{Background}}}
\]

(9)

Assuming that constant pressure can be applied and the ideal gas equation can be used to relate the molecular number density to the temperature of the flame, the following relation can be formed:

\[
\frac{T_{\text{Flame}}}{T_{\text{Air}}} = \frac{I_{\text{AirRayleigh}}}{I_{\text{Flame–Rayleigh}} L_{\text{Avg}} \sigma_{\text{Air}}}
\]

(10)

The only unknown that remains in Equation (10) is the flame shot cross section. The Rayleigh scattering cross section \(\sigma_{\text{Air}}\), which for a gas in vertically polarised light may be expressed according to Miles et al. [10]:

\[
\frac{\partial \sigma_{\text{Air}}}{\partial \Omega} = \frac{3 \sigma_{\Sigma} \rho_0}{8 \sigma_{\Sigma} (2 + \rho_0)}
\]

(11)

\[
\sigma = \frac{32 \pi \varepsilon (e-1)}{3 \lambda N} \left(\frac{6 + 3 \rho_0}{6 + 7 \rho_0}\right)
\]

(12)

The bracketed term in Equation (12) involving \(\rho_0\) is the King correction factor. Many investigators neglect the King correction factor term which introduces an error in each species cross section. An example of the possible error is if the King correction factor is ignored for CO, an error of up to 15% can be made in the cross section (Sneep et al. [14]). Due to values for the King correction factor being readily available (Sneep et al. [14]) and easily incorporated into the cross section calculation, we have included the King correction factor for species that the data is available.

Utilising 1D laminar premixed and strained opposed flow simulations using the open source Cantera code of Goodwin [5], the variation of cross section with respect to reaction progress variable has been determined. For the strained simulations the composition of the coflow combustion products is used for one stream and the composition of the central jet used for the other stream. A number of strain rates were evaluated, and a best fit polynomial evaluated for the change cross section with respect to Rayleigh signal. The polynomial follows closely the trend line even for the unstrained laminar case compared to the extinguished case. The average error comparing the strained and unstrained case to polynomial of best fit is around 6%.

3.2 OH Fluorescence image processing

In order to account for electronic quenching, vibrational transfer and fluorescence saturation a model needs to be utilised to relate the experimentally measured fluorescence intensity to the OH number density. Assuming a four level
model (Daily [3]) for the OH molecule, the rate equations for the populations in the electronically excited \( \Lambda \Sigma^+ \) \( ν=1 \) and \( ν=0 \) vibrational levels, \( N_4 \) and \( N_3 \) respectively may be expressed as:

\[
\frac{dN_4(t)}{dt} = -N_4 \tau_4^{-1} + B_{4} I_{\text{laser}} f_B(J^*)
\]

\[
\frac{dN_3(t)}{dt} = -N_3 \tau_3^{-1} + V_4 \tau_4 N_4
\]

Where \( \tau_4^{-1} \) and \( \tau_3^{-1} \) are the decay rates of populations of \( N_4 \) and \( N_3 \) respectively. \( B_4 \) is the Einstein absorption rate of the laser coupled ground state. \( I_{\text{laser}} \) is the laser photon density. \( C_{\text{OH}} \) is the number density of the OH molecule. \( f_B(J^*) \) is the Boltzmann fraction in the laser coupled rotational level \( J^* \) of the electronic ground state. \( V_4 \) is the vibrational transfer rate from \( ν=1 \) to \( ν=0 \). The \( N_4 \) and \( N_3 \) population decay rate constants may be expressed as:

\[
\tau_4^{-1} = A_4 + V_4 + Q_4 + I_{\text{laser}} B_4
\]

\[
\tau_3^{-1} = A_3 + Q_3
\]

Where \( A_4 \) and \( A_3 \) are the net spontaneous emission rates from \( ν=1 \) and \( ν=0 \). \( Q_4 \) and \( Q_3 \) are the electronic quenching rates from \( ν=1 \) and \( ν=0 \). \( B_4 \) is the Einstein spontaneous emission rate from the excited laser state to the laser excited ground state.

The excited OH lifetime in an atmospheric pressure hydrocarbon flame is reported to be less than 2 ns as reported by Köllner et al. [7] using direct lifetime measurement with picosecond excitation. The excitation system used was a seeded Nd:YAG pumping a dye laser, the typical output of which is a pulse of around 8-10ns duration. Based on the OH lifetime and the pulse duration it seems reasonable to assume that the system is in steady state for most of the pulse duration. By assuming a steady state population is established in \( N_4 \) and \( N_3 \) within the time scale of the laser pulse, the net rate of change of these populations may be assumed to be zero. Due to the use of a filter with a band pass centred at 314nm and a FWHM of 10nm the fluorescence intensity \( F \) will be primarily from the bands centered around 309nm and 314nm. Due to the high degree of attenuation from both the dichroic beam splitter and the 314nm filter at 280-285nm, we assume that no resonant fluorescence \( (N_4 A_{40}) \) or UV Rayleigh scattering is collected. The detected fluorescence may be expressed as:

\[
F \propto N_4 A_{42} + N_3 A_{31}
\]

Where \( A_{42} \) and \( A_{31} \) are the net spontaneous emission rates from \( ν=1 \) to \( ν=2 \) and \( \nu=0 \) to \( ν=0 \) respectively. Utilising Equations (10-13), Equation (14) may be expressed as:

\[
F \propto \frac{I_{\text{laser}} C_1 [A_{42}(A_{41} + Q_4) + V_4 A_{43} A_{31}]}{(A_{31} + Q_3) (C_2 + I_{\text{laser}} B_4 A_{41})}
\]

Where the constants \( C_1 \) and \( C_2 \) can be expressed as:

\[
C_1 = B_{4} C_{\text{OH}} f_B(J^*)
\]

\[
C_2 = A_{41} + A_{42} + V_4 A_{43} + Q_4
\]

Of particular interest from Equation (15) is that with the proposed fluorescence model, the fluorescence intensity seems to display an asymptotically correct relationship with the laser photon density, \( I_{\text{laser}} \). For small values of \( I_{\text{laser}} \) the \( N_4 \) loss term \( C_1 \) is much larger than \( I_{\text{laser}} B_4 \) term in the denominator, leaving a linear proportionality of the fluorescence to \( I_{\text{laser}} \), this is the well known “Linear Regime” of fluorescence. The regime of linear fluorescence is shown in Figure (1) for spectral energy intensities up to 300 MW/cm².

For infinitely large \( I_{\text{laser}} \) the fluorescence intensity will tend toward a steady value, similarly predicted by Lucht [8]. Also of importance is that Equation (15) predicts zero fluorescence when the laser power is zero.

Following from Equations (15-17) the mole fraction of OH \( X_{\text{OH}} \) may be expressed as:

\[
X_{\text{OH}} = \frac{F_{\text{OH}} \tau_3^{-1} \tau_4^{-1}}{\eta_{\text{OH}} I_{\text{laser}} B_4 f_B(J^*)} \left( \frac{P N_a}{R T} \right) \left[ 1 + A_{42} + A_{31} V_4 A_{43} \right]
\]

From Equation (18) \( \eta_{\text{OH}} \) represents the OH collection system through-put efficiency, \( F_{\text{OH}} \) is the dark noise corrected fluorescence signal, \( P \) is the pressure, \( N_a \) is Avogadro’s number, \( R \) the universal gas constant and \( T \) the corresponding temperature from the processed Rayleigh image. The spectroscopic constants necessary to process Equation (18) were taken from LIFBASE (Luque [9]).

![Figure 2](image-url)

Figure 2, a comparison of the normalised experimental and calculated LIF signal.

The experimental values used in Figure 2 were obtained by calibration in a uniform field of known OH concentration, provided by a nitrogen-shrouded laminar flat-flame burner Kelman et al. [6], running at stoichiometric conditions. At a point near the centreline of the UV beam, the fluorescence was monitored for a number of laser energies. By assuming a constant temperature and OH mole fraction field in the calibration burner, the calculated values were found by using the following simplified version of Equation (15):

\[
F \propto \frac{I_{\text{laser}}}{{\left( \frac{C_1}{{B_4 A_{41} + I_{\text{laser}}} \right)}}
\]

The excellent agreement of the experimental measurements and theoretical calculation in Figure 2 should be viewed as a verification of the functional form of the fluorescence to laser power in Equation (15), as well as the accuracy of the spectroscopic constants used for \( C_1 \) and \( B_{41} \).

In order to evaluate the electronic quenching rates from the excited \( \Lambda \Sigma^+ \) \( ν=1 \) and \( ν=0 \) levels, experimentally derived data for the two species dependant constants and the species quenching cross sections have been used. We use a similar
method to that of Chen et al. [1] to express the quenching and vibrational transfer rates:

\[ Q_3, Q_4, V_{d3} = \sum_{i=1}^{j} C_i a_i \sigma_i Q_i^{0.5} \]  

(19)

Where \( C_i \) is the \( i \)th species number density, \( \sigma_i \) is the species quenching cross section and \( a_i \) is species dependant constants. Paul [11] develops a relation to relate \( \sigma_i \) to the experimentally measured high temperature limit for the cross section \( \sigma_{Q_i} \), which can be expressed as:

\[ \sigma_Q = \sigma_{Q_0} \exp(e/kT) \]  

(20)

Using data for species electronic quenching and vibrational transfer from Tamura et al. [15] and Paul [12], the relation developed in (19-20) has been evaluated for a number of opposed flow simulations and curves fitted to the data, in a similar fashion to the Rayleigh data.

4. PROOF OF PRINCIPLE IN A TURBULENT FLAME

To illustrate the experimental method developed corresponding pairs of processed and unprocessed Rayleigh and OH images are presented in Figure 3 below. No attempt is made to draw any conclusions from these images. The images are for a coflow temperature of 1500K, central jet equivalence ratio of 0.5 and 100m/s average exit velocity. Images are taken at X/D=20 and each image size as presented is 25mm (W) x 11mm (H).

Figure 3, a) Raw OH image, b) Raw Rayleigh image, c) processed OH (mole fraction x 1000) and d) processed Rayleigh to temperature.

5. CONCLUSIONS

A quantitatively accurate method for evaluating 2-D Rayleigh and OH PLIF has been developed. Careful accounting in the 2-D Rayleigh signal for shot fluctuations in laser power, background reflections and variations in cross section has been compensated for. A model has been derived to evaluate the OH PLIF signal enabling quantitative evaluation of the OH concentration in both the linear and saturated regimes of fluorescence. The ability to utilise the non-saturated portion of the OH fluorescence permits much larger spatial areas to be examined with greater certainty. As a proof of principle, the developed experimental method is applied to a highly turbulent premixed flame, the PPVCB.

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REFERENCES