Stretch Rate Influence in Chemiluminescence Emissions of Pre-Mixed Laminar Flames

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ABSTRACT

The spatially resolved chemiluminescence emission of $\text{C}_2^*$ and $\text{CH}^*$ at the flame front of laminar Bunsen premixed propane/air flames was measured in this study. The flame compression and chemiluminescence intensity were studied in open air conditions for different equivalence ratios: 0.9; 1.0; 1.1; with a constant Reynolds' number of 1200. Local measurements of chemiluminescence were obtained using photographic recording, by a CCD camera, combined with the use of the algebraic reconstruction tomography technique. Stretch rate calculation was based on the location of $\text{C}_2^*$ maximum intensity emission varying from $-40 \text{ s}^{-1}$ to infinite, when the radius tends to zero, with small differences registered between the studied equivalence ratios. Chemiluminescence emission of $\text{C}_2^*$ and $\text{CH}^*$ was found not to be constant in the flame front due to heat, mass and species diffusion and flame compression. It was also found that flame compression decreases $\text{CH}^*$ signal maximum intensities, however the results obtained for $\text{C}_2^*$ showed that it does not depend of compression. The results indicated that the local intensities emission rate of $\text{C}_2^*/\text{CH}^*$ are not constant along the flame front and its value increases with flame compression.

Keywords: Laminar flames, Bunsen flames, Chemiluminescence, Tomography, Stretch rate

1. INTRODUCTION

Controlling the equivalence ratio inside a combustion chamber increases systems efficiencies, reduces pollutants emissions and also allows operating near its stable limits, by predicting combustion instabilities and preventing equipment destruction.

Thermocouples and microphones have been used to diagnose these oscillations, however due to the high temperatures achieved, that can damage the probes, and slow time response these equipment are some times, not a viable option. As an alternative, optical sensors based on chemiluminescence emissions offer the benefit of being able to collect information from extremely hostile environments in high frequency regimes. Since the application of these techniques is growing and becoming popular, it is important to understand the chemiluminescence mechanisms providing data for equipment calibration.

Chemiluminescence is the released energy in form of radiation by electronically excited molecules, when returning to their ground stage of energy. The origin and consumption of these radicals is related to the chemical kinetics nature of fuel oxidation, being $\text{OH}^*$, $\text{CH}^*$, $\text{C}_2^*$ and
CO₂* the easiest detectable molecules in hydrocarbons combustion. Each of these species is associated with photon emission in characteristic wavelengths, as it is possible to observe in Table 1.

According to this, several studies correlate light intensity, in the OH*, CH*, and C₂* wavelengths, to equivalence ratio. Kojima et al. [1] determined a power law correlation between air to fuel ratio, of a laminar premixed methane/air flame, with C₂*/CH*, CH*/OH* and C₂*/OH* emission intensities ratios. Ikeda et al. [2] obtained similar correlations to turbulent premixed propane/air flames. Equivalence ratio in liquid fuels combustion has also been studied [3], showing a linear dependency of C₂*/OH*, CH*/OH* and C₂*/CH* ratios with reaction stoichiometry. Several other combustion characteristics like pollutants emissions [4], heat released [5], flame temperature [6] have been correlated with chemiluminescence measurements as well. Despite of the important progress that previous studies gave to chemiluminescence comprehension, the results may only be valid with the burning system used.

This is due to simultaneous variations, while measuring light emissions, of equivalence ratio, stretch rate, temperature and pressure, invalidating the results to be extrapolated or to have universal conditions that relate a single parameter with the luminous signal. Even with this limitation, these techniques have been applied to practical control systems [8,9], gas turbines [10], internal combustion engines [11], where flame behaviour is notably turbulent and domestic low power laminar flames.

In this scenario of practical applications of the technique there are several factors that may influence the relationship between laboratorial and real flames. One of these factors is flame stretch, strongly present in turbulent and high curvature flames. Such flow characteristics interfere in chemical kinetics and may change the chemiluminescence mechanisms of formation and/or emission.

The stretch rate, $K$, defines stretch intensity as the rate of change of a flame surface element of area $A$, along the time [12] and it is due to aerodynamic efforts and flame curvature effects [13]:

$$K = \frac{1}{A} \frac{dA}{dt}$$

Applying the flame surface equation, $G$ function, this general expression for flame stretch rate can be simplified for the case of axisymmetric geometry, where Bunsen flame is included. Adopting the cylindrical $(r, \theta, z)$ coordinate, it yields [13]:

<table>
<thead>
<tr>
<th>Molecules</th>
<th>Wavelength [nm]</th>
<th>Life time [s]</th>
</tr>
</thead>
<tbody>
<tr>
<td>CH*</td>
<td>431.5</td>
<td>$5.6\pm0.6\times10^{-7}$</td>
</tr>
<tr>
<td>C₂*</td>
<td>416; 466; 516,5 e 566</td>
<td>$1.7\pm0.2\times10^{-7}$</td>
</tr>
<tr>
<td>OH*</td>
<td>306.4</td>
<td>$1.5\times10^{-6}$</td>
</tr>
<tr>
<td>CH₂O*</td>
<td>395; 423</td>
<td>continuous emission</td>
</tr>
<tr>
<td>CO₂*</td>
<td>continuous emission</td>
<td></td>
</tr>
</tbody>
</table>

Table 1: Excited radicals characteristics [7].
evaluated at the flame surface. This result is general in that axial flow velocity, $w$, and Bunsen flame angle, $\alpha$, can be general functions of $r$ and $z$. If $w$ and $\alpha$ are assumed to be constants such that the flame surface is a circular cone with sharp apex, then [13]:

$$K = -\frac{V_0 \sin 2 \alpha}{2 R_f}$$  \hspace{1cm} (3)$$

Note that the stretch for the axisymmetric Bunsen flame is negative, indicating that the Bunsen flame actually suffers compression and its intensity increases to infinite with decreasing flame radius $R_f$, as the flow moves towards the apex.

Several are the studies that show the stretch rate influences in combustion reactions. Law and Sung [13] studied the effect of stretch in laminar flame speed, heat released and maximum temperature, in stationary planar, spherical and Bunsen geometries, concluding that for conditions far from flame extinction, the value of these properties increase with increasing $K$, for Lewis’ numbers smaller than unity (lean methane and rich propane), and decrease for $Le > 1$ (rich methane and lean propane). Marley and Roberts [12] verified, for spherical propane/air flames, similar results, showing that laminar flame speed, $S_L$, decreases with increasing stretch rate, for equivalence ratios less than 1.34, and the opposite for greater values.

In relation to light emissions, Hardalupas and Orain [10], using a counterflow burner, supplied with natural gas and a Cassegrain optics, showed the independence of the OH*/CH* relation with stretch rate. On the other hand, a strong relationship with the C$_2$/CH* signals ratio and stretch rate was verified for rich mixtures.

In order to better understand the influence of stretch in chemiluminescence it is important to evaluate, locally, light emissions along a flame geometry subjected to a high curvature where stretch intensity changes, as it is the case of axisymmetric Bunsen flames.

By using axisymmetric configurations it is possible, with the combined use of computerized axial tomography (CAT) and integrated images of the entire flame, to locally measure chemiluminescence emitted from the reaction zone. This technique has been applied by several authors to measure such parameters as: flame temperatures [14], pollutants location and formation [4] and heat released [15].

The purpose of this work is to assess chemiluminescence emissions of CH* and C$_2$*, in a Bunsen propane/air flame, under compression influence, using tomographic reconstructions.

On the second chapter it is described the experimental set up, the CAT algorithm used, the flame stretch calculation and an error analysis is carried out. The results are discussed in chapter three. Finally the work is summarized with conclusions on chapter four.
2. EXPERIMENTAL SET UP

To infer the influence of stretch in chemiluminescence emissions, light intensity measurements were conducted, using tomographic reconstructions of the integrated reaction zone, at the wavelength emissions of CH* (431.5 nm) and C2* (516.5 nm), in Bunsen propane/air flames, at atmospheric pressure, for a Reynolds number of 1200 and an equivalence ratio range of 0.9 to 1.1.

The experimental arrangement used is presented in Figure 1 and is composed by the burning and the optical acquiring systems.

A convergent nozzle was used to stabilize Bunsen flames, in a range of 0.9 to 3 kW of power output. The burner has an 18 mm exit diameter and a contraction ratio areas of 6.53. The nozzle was attached to a decoupling unit that consisted of either three discs, each 5 mm thick, made of packed glass spheres and equally spaced, through which the fuel/air mixture is injected.

Both air and propane flows were controlled by two Alicat Scientific, 16 Series, electronic flow meters with a reading error of \( \pm (0.008 \dot{V} + 0.002 \dot{V}_{\text{max}}) \) 1/min, with \( \dot{V}_{\text{max}} \) of 20 l/min and 5 l/min for air and propane respectively.

The image acquisition was carried by a 2D spatially resolved system, where spectral range selection was made by placing two Melles Griot interference filters in front of the lens. This allowed to capture CH* (431.5 nm) and C2* (516.5 nm) signals, with 11.02 and 9.14 nm bandwidth and a transmissivity of 44.31 % and 55.47 %, respectively.

A Cosmicar/Pentax photographic lens, with a 12.0 mm maximum focal distance and a relative aperture of 1:12 visualized the entire reaction zone, converging the image into a CCD sensor of an uEye camera. The analogical luminous signal was then converted to an electrical signal allowing to reproduce the image digitally, in the RGB color model, in which the square side of a pixel corresponds to 48.3×10^{-3} mm. Such parameters as pixel clock, exposure time, master gain, red, green and blue gain and black level offset were set up using the commercial IDS software.

**Tomography**

This spatially resolved technique is based on the mathematical conversion of lateral measurements series of spectral radiation into radial profiles of radiation.
Assuming that an axisymmetric flame may be modelled as a ring set of constant scalar properties (monochromatic radial emission coefficient, $I_\lambda$, in the present study), the integrated signal $J_\lambda$ is an implicit radial function given by:

$$J_\lambda(x) = 2 \int_x^\pi \frac{I_\lambda(r) r \, dr}{\sqrt{r^2 - x^2}}$$

(3)

Using Abel’s integral inversion [14] it is possible to know the $I_\lambda$ values:

$$I_\lambda(x) = -\frac{1}{\pi} \int_x^\pi \frac{\partial J_\lambda(x) r \, dr}{\partial x} \frac{1}{\sqrt{x^2 - r^2}} \, dx$$

(4)

Equation (5) presents the radial emission coefficient as a function of geometry and radiance lateral measurements distribution.

When assuming a real Bunsen flame as an axisymmetric geometry, it has the advantage of that a single reaction zone projection supplies all the information needed for its reconstruction, reducing computational time, as well as an economy of experimental equipment, since it is only one camera needed. The physical principle of tomography application is shown at Figure 2.

However, for the practical application of equation (5), it is needed its discretization using numerical methods described on [14].

Algebraic reconstruction may be used alternatively to Abel’s integral inversion as a tomography algorithm. In this formulation the integrated intensity values detected in each zone, $J_{ki}$, are calculated by summing all the intensities of each ring, $I_j$, and multiplied by the resultant area $A_{ij}$ of interception between probe visualized zone and the constant ring properties [16].

This methodology application results of the projection discretization into a $2n \times l$ mesh, in which as more image elements are used, the better results will be. Figure 3 illustrates the method applied to a generic Bunsen flame, where a) represents the mesh and b) the tomography execution applied to the element line $k = 1$.

In this process there are $n$ zones and $n$ rings, in which $J_{ki}$ value represents the integrated signal, detected by the probe along the zone $i$. The method is mathematically expressed by equation (6), which developing yields on equation (7).

Knowing the $J_{ki}$ and $A_{ij}$, it is possible to determine $I_j$ values allowing the reconstruction of the original flame.

The program developed to implement the algebraic reconstruction algorithm is the “Tomograf_6c”, adapted from the “Tomograf_5” code.
In this program each pixel corresponds to an element. The collected data is converted from RGB to grayscale images, in which the gray level is a function of the monochromatic radiation intensity.

Since the arrangement of a Bunsen flame is assumed to be axisymmetric in geometry and properties, the algorithm determines the centre of flame by a method akin to the determination of the centre of mass in a body. \( A_{ij} \) areas are calculated based on the image, using as unities of measurements the number of pixels. Equation (6) is now possible to solve and the reaction zone is reconstructed in a light profile form.

By using this technique it is not only possible to identify the flame front in axisymmetric geometries, but also assess chemiluminescence intensity along the reaction zone.

**Stretch Rate Calculation**

Knowing the \( \text{CH}^* \) and \( \text{C}_2^* \) signal intensities it is needed to assess flame stretch conditions.

Since it is not possible to determine analytical \( K \), in practical Bunsen flames, an algorithm was developed to calculate its numerical value. This method uses the tomographic reconstruction to locate, in a referential centered with the flame, the points in which maximum chemiluminescence intensity occurs for every pixel line. Through each of this points and using its eight neighbors, a linear regression is plotted, in which the slope defines the local \( \alpha \) angle.

Assuming flow axial velocity as constant and applying central differences to equation (2), it is possible to estimate the local stretch rate value in real Bunsen flames.
Uncertainties analysis

In order to validate the experimental results, errors related with measuring equipment and used techniques are presented.

In image acquisition the colour scale was calibrated using the IDS software, defining the black level offset.

To determine electronic noise intensity, photographic recordings were performed with the system insulated from the surrounding radiation. It was verified that interferences imposed by equipment were zero for all experimental set ups. Also, the error derived from the environmental noise was determined, being less than 0.8 % and having no relevance in the obtained results.

To quantify the error associated with the tomography algorithm, Correia et al. [17] proceeded to the reconstruction of mathematical generated forms, concluding that the differences were mainly due to the projection discretization, with an associated error below 5 %. It is also important to refer that this process is only valid in the absence of radiation auto absorption by the surrounding gas [17]. However, Gaydon and Woflhard [18] and Porter et al. [19] showed that measurements based on C2* and CH* radicals are not affected by auto-absorption in non luminous flames.

For the stretch rate calculation, linear regression based on nine experimental points was used, with a progressive refinement when approaching towards base and the tip of the flame. The choice of nine points was a compromise between the minor relative difference, between mean $\alpha$ and RMS values, Table 2, and image tip resolution (20×20 pixels), avoiding the insensitivity to angle variations that occur in this region.

<table>
<thead>
<tr>
<th>Number of data points used for line regression in flame front</th>
<th>3</th>
<th>5</th>
<th>7</th>
<th>9</th>
</tr>
</thead>
<tbody>
<tr>
<td>Relative Difference $</td>
<td>\alpha_{\text{mean}} - \alpha_{\text{RMS}}</td>
<td>$</td>
<td>77.25 %</td>
<td>44.56 %</td>
</tr>
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</table>

Table 2: Relative difference between mean $\alpha$ and RMS values in a Bunsen flame stretch rate.

3. RESULTS

The stability diagram of the burning system is presented on Figure 4 with typical Bunsen flame modes photographically reported on Figure 5. Stability diagram shows the flash back and blow off limits, along several values of equivalence ratio and Reynolds’ numbers.

Based on Figure 5 it is possible to observe the color and intensity differences of light emitted for the different mixture proportions. Light is
more intense and green in rich flames and more dim and blue for lean ones.

It can also be verified that for the same Reynolds’ number, flame height decreases with equivalence ratio towards to stoichiometry conditions, returning to increase with the increasing mixture richness. That can be explained by the fact that the laminar flame speed is varying and its maximum being located at $\phi = 1.1$ [12].

Photographic recordings, using interference filters and tomographic reconstructions were performed, allowing to quantify chemiluminescence emissions, for different equivalence ratios. Figure 6 a) presents the flame spatially integrated image in the wavelengths emission of $C_2^*$ and $CH^*$ and their CAT outputs on b). On Figure 6 b) each figure is composed by the tomographic reconstruction on the left and the original image reproduced in grayscale on the right side.

Flame stretch calculation is presented on Figure 7, where circles represent the experimental results calculated based on numerical application of equation (2); the squares corresponds to the mean values of $K$ in intervals of $R_f/r = 0.02$; and the continuous line is the theoretical result given by equation (3). From these graphics it is possible to confirm that the theoretical solution
agrees with the experimental one.

In all the three cases, flames are subjected to compression (negative stretch) and presents low values of $|K|$, in a range of 40 to 200 s$^{-1}$, for the constant slope zone (between $R_f/r$ values of 0.2 and 1.0). Moving towards the tip compression increases very fast tending to infinite when $R_f/r = 0$.

It is also important to mention that flame stretch varies with equivalence ratio. As it was discussed before, laminar flame speed depends on $\phi$ having its maximum at equivalence ratio of 1.1, resulting in lower flame height for $\phi = 1.0$ and 1.1 with a more open $\alpha$ angle. However, these are small differences that can not be directly observed from Figure 7. Figure 8 shows these differences for $R_f/r$ range of 0.2 to 1.0.

Figure 9 and Figure 10 show the spatial profiles of $C_2^*$ and $CH^*$ intensities, respectively, for the same three equivalence ratios. Again, circles represent the experimental data and the squares the mean values for intervals of $R_f/r = 0.02$ for $0.02 < R_f/r < 0.20$ and intervals of $R_f/r = 0.05$ for the remain flame. Data between $0.00 < R_f/r < 0.02$ were ignored, due to the uncertainties resulted from the high frequency oscillation of the flame tip.

The variation of $C_2^*$ emissions with equivalence ratio agrees with the results obtained by Kojima et al. [20], with methane-air flames, where for the range of equivalence ratios experimented signal intensity increases with increasing $\phi$. On the other hand $CH^*$ emissions appears to be less sensitive to the equivalence ratio variations.

The profiles of chemiluminescence emissions of $C_2^*$ specie present three distinct zones, for these three equivalence ratios: A first zone, A, $(0.80 < R_f/r < 1.00)$, where $C_2^*$ signal decreases; a second zone, B, $(0.20 < R_f/r < 0.80)$, with an almost constant signal intensity, exception made to $\phi = 1.0$; and a third one, C, $(0.02 < R_f/r < 0.20)$, where light intensity decreases again. The same three zones are also verified in the $CH^*$ profiles, however with differences on second zo-
Figure 7: Experimental and theoretical Bunsen flame stretch rate, measured with \( C_2^* \) emission: a) \( \phi = 0.9 \); b) \( \phi = 1.0 \); c) \( \phi = 1.1 \).

Figure 8: Theoretical Bunsen flame stretch rate for equivalence ratios of 0.9, 1.0 and 1.1.

The profiles registered on zone A are due to heat losses to the burner ring, cooling the flame, when approaching to the anchor point. Also the local equivalence ratio differs from the rest of the reaction since air and air/fuel are mixing due to flow momentum and mass diffusion, making the mixture locally leaner [6] and interfering in the chemiluminescence mechanisms.

In the centre of the flame, zone B, where the mentioned heat and mass transfer mechanisms have less expression on chemiluminescence emissions, \( CH^* \) intensity decreases when moving towards the tip, however \( C_2^* \) intensity appears to be more constant, along flame front. This \( CH^* \) signal intensity decreasing may be due to the increasing compression that the flame is exposed to. Figure 11 and Figure 12 were obtained by plotting the theoretical stretch rate versus the section mean values of chemiluminescence intensity. Observing them is
Figure 9: C2* maximum intensities emission along Bunsen flame front: a) $\phi = 0.9$; b) $\phi = 1.0$; c) $\phi = 1.1$.

Figure 10: CH* maximum intensities emission along Bunsen flame front: a) $\phi = 0.9$; b) $\phi = 1.0$; c) $\phi = 1.1$. 
Figure 11: C2* intensity as a function of stretch rate in Bunsen flames.

Figure 12: CH* intensity as a function of stretch rate in Bunsen flames.

easy to conclude that for $|K|$ values between 50 and 200 s$^{-1}$, that corresponds, approximately, to $R_f/r$ limits of zone two, C2* behaviour is inconclusive and CH* signal intensity actually decreases. The tendency observed on zone C may be explained by the tip concave geometry that concentrates large amount of heat and species [13], overcoming the compression effect of reducing chemiluminescence intensity and increasing the C2* and CH* signals.

Excluding the anchor and tip zone, where transfer mechanisms prevail, it is possible to conclude that stretch rate has a greater influence on CH* emission than on C2*.

The quotient of different species, quite used to determine the equivalence ratio of pre-mixed flames [1, 2, 10], becomes more accurate to determine $\phi$, being more sensitive to small variations of C2* and CH*. Intensity ratio profiles of C2*/CH* are shown on Figure 13, in which chemiluminescence emission ratio showed not to be constant in the all three zones, as previously defined. In the second zone, where correlations have been sketched to assess global equivalence ratio in Bunsen flames [2], the local emission ratio decreases with radius increase. The present work does not invalidate previous studies that show a constant value of C2*/CH*, since different flame geometries as well as collecting light systems and techniques used visualizes different flame volumes, which have a big influence on the results, however, applying those correlations to local emissions may suggest inconsistent equivalence ratios.

Figure 13: C2*/CH* intensities ratio along Bunsen flame front.
On Figure 14 it is possible to see the relationship between chemiluminescence intensity ratio and stretch rate, in which $C_2^*/CH^*$ increases with the increase of $|K|$, for zone B. The relationship between $C_2^*/CH^*$ and $\ln(|K|)$, in this zone, can be described as a linear function, in which the slope increases with equivalence ratio. It is also possible to see that on the tip zone, C, this relationship seems to tend to a constant value, when compared to zone B. Based on collected chemiluminescent signals, determination of $\phi$ using $C_2^*/CH^*$ rate depends on where the measurement is made. Even the calibration procedure, generally obtained using global signals, must be used with precaution, when applied to laminar or turbulent flames, under compression effect, since signals rate vary, besides other parameters, with stretch rate along flame front.

### 4. CONCLUSIONS

Spatially and spectrally resolved chemiluminescence emissions of a premixed laminar Bunsen propane/air flames were measured in open air conditions for different equivalence ratios, ranging from 0.9 to 1.1, with a constant Reynolds’ number of 1200. The spatially resolved chemiluminescence measurements of $C_2^*$ and CH$^*$ species were successfully performed at the flame front for different equivalence ratios using a CCD camera and a tomography algorithm. Flame stretch was also successfully determined using the $C_2^*$ maximum intensities location. Stretch rate in Bunsen flames is negative, which indicates that the flame is actually exposed to compression and varies from $-40 \text{ s}^{-1}$ to $-200 \text{ s}^{-1}$, where tip curvature has no influence, and from $-200 \text{ s}^{-1}$ to infinite in the limit when $R/r \to 0$.

Three distinct zones of chemiluminescence emissions were defined, for Bunsen flames, in which chemiluminescence emission profiles differs due to the different phenomenon present. A first zone, close to the burner ring, is influenced by heat losses to the cold metal of the burner and a local equivalence ratio leaner than the remain reaction; A second zone in which CH$^*$ emission is mainly affected by flame compression; and a third zone, near the tip, where the reaction is heated and has a large species concentration.
The relationship between $C_2^*$ emission and stretch rate is not monotonic. However, it is possible to conclude that flame compression has an influence on chemiluminescence mechanisms of $CH^*$, making their intensities decrease with increasing $|K|$. Local signals intensity ratio of $C_2^*/CH^*$ also varies along the second flame zone, increasing with the increase of $|K|$. 

REFERENCES