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A comparison of experimental results of soot production in laminar premixed flames

Abstract: Soot emission has been the focus of numerous studies due to the numerous applications in industry, as well as the harmful effects caused to the environment. Thus, the purpose of this work is to analyze the soot formation in a flat flame burner using premixed compressed natural gas and air, where these quasi-adiabatic flames have one-dimensional characteristics. The measurements were performed applying the light extinction technique. The air/fuel equivalence ratio was varied to assess the soot volume fractions for different flame configurations. Soot production along the flame was also analyzed by measurements at different heights in relation to the burner surface. Results indicate that soot volume fraction increases with the equivalence ratio. The higher regions of the flame were analyzed in order to map the soot distribution on these flames. The results are incorporated into the experimental database for measurement techniques calibration and for computational models validation of soot formation in methane premixed laminar flames, where the equivalence ratio ranging from 1.5 up to 8.

Keywords: soot volume fraction; light extinction technique; premixed flat flame

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1 Introduction

Soot can be defined as the solid particulate matter produced during combustion [1]. The process of formation is based in the conversion and agglomeration of the carbon atoms contained in the hydrocarbon fuel molecules. The formation of soot is not completely understood due to the complexity and velocity of this transformation. Soot is of great interest in engineering, insofar as its formation increases the radiated heat transfer, which can be a positive to the considered system. The excessive presence of particulate in diesel engines is, an example, where soot is harmful. These particles resulted from an inadequate combustion process can become logged in the seat of valves, preventing a proper seal in the combustion chamber and leading to a loss of compression in the cylinder. Another example is gas turbines, where the presence of soot affects the life of the blades due to abrasion for the contact.

The emission of this type of particulate matter into the atmosphere causes great harm to the environment, since there is a significant deterioration in air quality. Also, induce respiratory problems in humans and is associated with the occurrence of cancer. On the other hand, in industrial heating equipment such as a boiler, the presence of soot increases the heat exchange by radiation from the flame to the system walls, whereas the solid material resulting from the process absorbs and emits radiation at a wide range in the wavelengths spectrum [2]. Thus, where a large and uniform heat transfer is required, the presence of soot in combustion is purposely induced.

To control the emission of soot in real combustion systems, it is necessary to understand the soot formation and oxidation mechanisms. The basis of knowledge is the development of models that can predict the particulate matter formation, and the validation of these models requires information about the soot volume fraction. In this way, optical techniques are generally employed to perform the measurement of these quantities due the non- intrusive probing.

The necessity for such information in the study of combustion was what motivated the present work, which was developed in order to produce significant results for future calibration of measurement techniques and validation of theoretical models in this subject. Hence, the main objective of this study is to analyze the production of soot in premixed laminar flat flames of natural gas and air, measur-

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ing the soot volume fraction applying laser light extinction technique in different flame heights for equivalence ratio of 1.5 up to 8, finally, comparing the results with the literature.

2 Background

The effects of light scattering by particles have been investigated for more than a century, starting in 1861, even before Maxwell proposed the theory of electromagnetism. Lord Rayleigh applied Maxwell's equations and formally established the theory of Rayleigh scattering in 1881. Other notable scientists who contributed to this line of research include Lorenz in 1890, Mie in 1908, and Debye in 1909. These last two extended the Rayleigh theory to incorporate large and non-spherical particles. Previous studies applied the Rayleigh theory to monitor the mass concentration of the particulate of different sources, such as diesel engines. In the last two decades, several researchers have shown results on the extinction coefficient from studies using hydrocarbon fuels subjected to various combustion conditions. These results are validated based on the use of the light extinction technique and a resume of them was made by Mulholland [3].

The light extinction technique is used in the region of the flame itself, as well as in regions outside of it. For situations that include the flame, the measurement of the soot volume fraction is the main information that helps to understand the development of the particulate material in the combustion. Moreover, the measurement of soot volume fraction by methods such as light extinction is of fundamental importance in the study of radiation transport in combustion.

The determination of soot volume fraction based on the light extinction technique with laser light was performed previously [4]. This method is broadly employed by the scientific community that investigates combustion, since the results obtained are backed up by non-intrusive measurements and generate instant and useful information.

Several optical measurement techniques are calibrated using this method, for instance, the Laser Induced Incandescence method, LII. It has been used to validate the soot volume fraction obtained based on the LII 2D technique in combustion systems with laminar diffusion flames. The LII technique presented dispersion in the range of 5 to 10%, probably due to the difference in particle size [5].

Other researchers contest the findings in the literature for the refractive index of soot particles, which is a key parameter in determining the extinction coefficient [6]. Measurements were performed for the wavelengths of 450, 630 and 1,000 nm using crude oil as fuel. Results have shown that the extinction coefficient for the three wavelengths considered is constant, even though the particle number concentration varied by a factor of 24 due to agglomeration.

Measurements of acetylene premixed flames using both light extinction and gravimetric techniques were performed to obtain the soot volume fraction [4]. After calibration, an extinction coefficient of 8.6 was found and the author states that this number can be applied as the extinction coefficient for soot generated from the combustion of various fuels for lighting systems that include the visible spectrum. The conclusion is that simultaneous measurement methods must be employed for more accurate results of the extinction coefficient.

The light extinction technique was also used by in order to verify the formation of soot into a diesel engine cylinder [7]. The results were compared with other techniques such as the LII and emission in two colors methods to obtain an extinction coefficient with less uncertainty.

Studies on the influence of oxygen and air velocity on soot formation employed the technique of laser light extinction in a diffusive acetylene flame with an annular coaxial injection of oxygen and air [8]. Oxygen enrichment produces an increase in soot formation, as the use of higher injection velocities of air in comparison to fuel. Thus, a control of the amount of oxygen injected and the air velocity can be useful tools for the control of soot production.

Data of the concentration of soot in a non-premixed turbulent ethylene flame were obtained by applying light extinction and scattering technique [9]. The measurements of soot volume fraction in several positions of the flame were performed in the radial and axial direction. A maximum soot volume fraction of 1 ppm was found among all measurements.

The light extinction technique was also employed to obtain quantitative values of the soot volume fraction in sprayed biodiesel flames [10]. LII measurements were made using the light extinction technique results for calibration, which showed a maximum 10% discrepancy between the results.

Furthermore, numerical models can be validated by experimental results measured using the light extinction technique. Authors such as [11, 12] used this technique to validate theoretical models of soot formation using methane and acetylene, respectively.

3 Methodology

The transformation of fuel in soot after combustion takes place in five steps, as shown in the Figure 1. The fuel pyrolysis forming the precursors through the processes of cracking, coking and cyclization, in order to form polycyclic aromatic hydrocarbons (PAH), which are composed of ethane (C_2H_6). In sequence the Nucleation occurs when a large molecule precipitates, forming a solid particle which shape is considered as a sphere. In the coalescence process, two particles unite to form a larger particle. The surface grows up when the particle absorbs other components, further increasing the size. Finally, the agglomeration is the union of several of these previously formed particles, which results in a solid conglomerate [13].

The laser light extinction technique is based on the Beer-Lambert Law which is an empirical relationship that relates the absorption of light to the medium properties that it passes through. Thus, measurements of light intensity are performed without flame and with laser passing through the flame, which the particles of soot in the laser line of sight attenuate the initial intensity. When applied to the light extinction technique for regions within the flame, becomes [4]:

$$\frac{I}{I_0} = \exp\left(\frac{x f_v k_e}{\lambda}\right). \qquad (1)$$

The approach consists in a burner, the light source, detection and control systems, as shown in the Figure 2. This apparatus was constructed in order to ensure alignment between the components of the illumination and the detection system. The flowfield was characterized by the similar way as done in previous works [14]. As result, a planar and adiabatic flame is obtained.

Two flowmeters were used to control the air and fuel velocities in order to establish the equivalence ratio of the mixture for each flame studied. The gas line from the CNG (Compressed Natural Gas) tank was in 4 bar of pressure and the compressed air line at 3 bar.

The flowmeter used to measure the fuel flow has a scale from 0.2 to 2.0 $Sl.min^{-1}$ and another to air ranging from 0.001 to 0.01 m^3/s, both of Conaut 420, with 5% of uncertainty.

For the detection system a photomultiplier tube was used as photodetector, Hamamatsu, model 931 A. The device has a quantum efficiency of approximately 3.75%, with 2% of uncertainty, for the wavelength of 532 nm emitted from a laser diode, continuous wave, of 1,000 mW power, with an uncertainty of 10 mW.

The photomultiplier tube was connected to a high voltage source, Fluke, model 415B, which provides a voltage of 1,000 V to amplify the signal intensity. The signal is captured by a digital datalogger multimeter, Agilent, connected to a microcomputer. The HP BenchLink Data Logger program is responsible for the interface, which enables capture of the electrical signal from the photodetector.

A slit was used to prevent the light of the flame to noise in the signal. An optical bandpass filter centered at 532±25 nm was used for further reducing the transmission of ambient and flame light. Thus, only the laser light inside on the photodetector. Finally, a cylindrical lens was positioned after the filter to allow the entire area of the photodetector to be illuminated homogeneously.

The study was conducted in a burner designed in previously studies [15]. The burner base allows the homogeneous mixture of air and fuel, which exits through a disc of 32 mm in diameter with 1,639 holes of 0.5 mm, arranged in a hexagonal pattern. A thermal bath provided water at 85°C to traverses the entire perimeter of the perforated disc in order to avoid the heat exchange between the flame and the burner body, ensuring adiabatic conditions at the base of the flame. This setting allows the development of flat, i.e., one-dimensional planar flames.

The measures were filed in spreadsheet format, which were processed using the program iWork Numbers, from Apple, in order to process the results to yield the f_v values.

The fuel used in the burner was CNG, which can be easily found at gas stations. According to the supplier, this gas consists of 91% CH_4 (methane), 6.1% of C_2H_6 (ethane), 1.1% of C_3H_8 (propane), 0.4% of CO_2 (carbon dioxide) and 1.4% of N_2 (nitrogen). The calorific value of CNG is around 38.8×10^6 J/m^3.

Table 1 presents the flame settings used in the experiment. The flow rates are adjusted to the air pressure and fuel lines, 3 and 4 bar, respectively. For all flame configurations used, the Reynolds number was below 2,100 that is the upper limit for laminar flows.

The light extinction technique is based on measuring the intensity of the laser light that passes through a flame, where part of the light is absorbed, scattered and attenuated due to the presence of particulate material. Therefore, to measure the intensity of transmitted light, I. The burner was positioned on a support, between the illumination and the detection system, to allow measurements at different heights. The first measurement was performed at 5 mm above the base of the flame. Subsequent measurements were made at intervals of 5 mm. Measurements of I_0 were obtained in the same way as for I, with the difference that the laser light focused directly on the photomultiplier tube, without crossing the flame.

The field of view was spatially calibrated using images of the burner nose these images were processed with

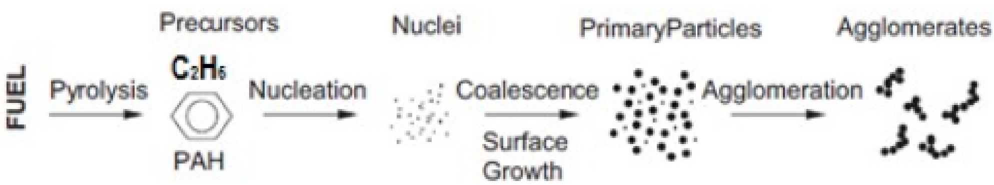

Figure 1: Stages of the soot formation process [13].

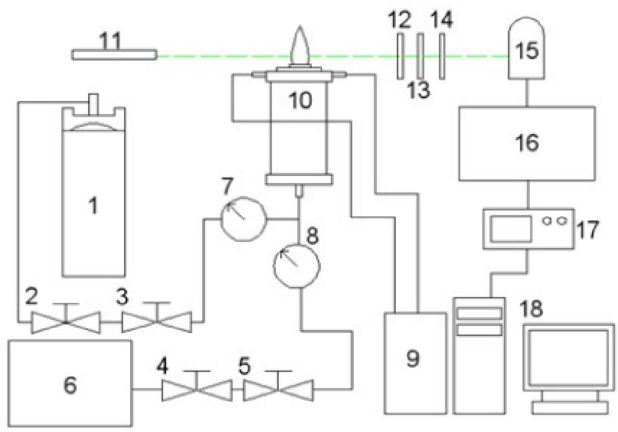

Figure 2: Scheme of the experimental apparatus. 1) Fuel vessel, 2, 3, 4 and 5) Throttle pressure and flow, 6) Air compressor, 7 and 8) Flowmeters, 9) Thermal bath, 10) Burner, 11) Diode laser, 12) Slit, 13) Band-pass optical filter, 14) Cylindrical lenses, 15) Photomultiplier tube, 16) High Voltage Supplier, 17) Datalogger Multimeter, 18) Computer.

Adobe Photoshop CS5 software. The flame thickness, at each height where measurements were made, was measured using the comparison with a known real dimension present in the image. The process consists in comparing the number of pixels that make up the flame with the number of pixels of a pattern with known size at the picture.

Thus, from the data of I and I_0 obtained experimentally, the knowledge of x and considering the assumption of $k_e = 8.6$ [4], values for f_v could be obtained for the chosen points of the flame and for the different equivalence ratios as detailed in Table 1.

Soot particle size was considered smaller than the laser light wavelength. Thus, the flames configuration is in the Rayleigh regime, where the absorption section is much larger than the scattering one, which allows the applicability of the light extinction technique. Moreover, the light absorption occurs primarily by soot particles, not being verified for combustion products [16].

The values of I and I_0 are given by the electric signal which is measured by the datalogger, in terms of voltage. These results were submitted to the Chauvenet criterion to identify values that go beyond the mainstream trend. The criterion specifies that a measured value can be rejected if the probability of obtaining the deviation from the average is less than $0.5n$. Where n is the number of measurements for each measured equivalence ratio.

3.1 Measurement uncertainty

The analysis of experimental uncertainty (u) caused by random errors, which lies in result of the addition of small independent and uncontrollable errors, such as errors of measuring instruments, was performed by applying the method of propagation of independent errors proposed by Kline-McClintock.

This method was applied to the results of soot volume concentration. For the error regarding the measurement of the flame width crossed by the laser light was considered as $u_x = \pm 3 \times 10^{-6}$ m. This value is consistent with the uncertainty in measuring the pixel size during the flame images analysis. The uncertainty of the light extinction factor was considered $u_{ke} = \pm 1.5$ [4]. The uncertainty for the average of values obtained in the light intensity acquisition were considered directly of the electrical signal, in *Volts*, as $u_{I0} = u_I = \pm 0.5\%$. It were made 600 measurements for each ϕ and for each height above the burner considered in this work. In this way, the value was obtained from the ratio between the largest value of the measured RMS signal amplitude, 200 mV, and the average of this signal, 1 mV, where the major ratio value in all measurements was considered. The uncertainty related to the wavelength emitted by the laser was considered $u_\lambda = \pm 10$ nm, from manufacturer information. Thus, the maximum uncertainty in the measurements of f_v is about 8% for the results of this work.

A similar analysis was made for the equivalence ratio uncertainties, using the associated of flowmeters uncertainties, that is 5%. Therefore, the uncertainty associated with the air flowmeter to the largest flow used was 0.2 Sl.min^{-1} and with the fuel 0.15 Sl.min^{-1}. Thus, the measurement uncertainty in the equivalence ratio values of the air/fuel mixtures is approximately 7%.

Table 1: Flames configurations.

Flame case	$\acute{V}_{air}$	$\acute{V}_{comb}$ $\acute{V}_{fuel}$	ϕ	R_e	$U_{global}(m/s)$
1	0.0083	0.0011	1.5	708	0.49
2	0.0083	0.0015	2	736	0.51
3	0.0083	0.0018	2.44	765	0.53
4	0.0083	0.0026	4	823	0.57
5	0.0083	0.0035	5.3	890	0.62
6	0.0083	0.0053	8	1,024	0.71

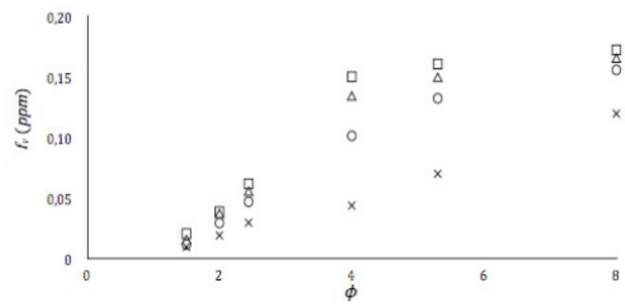

Figure 3: Relationship between the f_v formed for each ϕ used. The following symbols represent the measuring height from the base of the flame: (x) of 5 mm, (o) of 10 mm, (△) of 15 mm and (□) of 20 mm.

4 Results and discussion

The values of f_v were obtained on different conditions, presented in the Table 1. Measurements were taken at several heights, h, starting at the flame base. The measurements were not possible at $h = 0$, due to the low soot concentration. This corroborates the fact of having a quasi-stoichiometric region.

4.1 Behavior of f_v as function of the equivalence ratio and flame height

The behavior of f_v as function of the equivalence ratio and flame height is shown in the Figure 3. Values of ϕ smaller than 1.5 are considered critical (ϕ_c) because there is virtually no soot, according to the literature [12, 17]. Therefore, f_v measurements were performed for $\phi \geq 1.5$.

In flames with equivalence ratio of $2 < \phi < 4$ was observed a sharp increase in the values of f_v. This increase was greater as higher heights h were taken for the measurement. This behavior was expected and is consistent with the results presented previously. This occurs due to excessive quantities of fuel in the flame whose conditions are suitable to converting the carbon molecules contained in the gaseous fuel into soot particles. For values of the equivalence ratio higher than $\phi = 4$ there is a reduction in the growth rate of f_v, as observed for height measurement, but not for an oxidation reason. This behavior is attributed to the flame settings, which do not reach enough power, area and temperatures for the production of soot. The rest of the fuel is released in the form of unburned gases in the exhaust.

The statement about the decline in the growth of f_v for very rich flames was based on the burner characteristics and on the smell from the fuel present during the measurements for these flame configurations and by a toxic gas detector. A gas analyzer or fuel mapping technique would be useful to confirm this statement, but such alternatives were not available in the moment of the measurements.

The f_v values presented an increasing variation throughout along the flame height, beginning in the base from which the measurements were taken. Stronger growth of f_v was noticed until $h \approx 15$ mm, where the amount of soot increases up to 300% for equivalence ratios above 4. Thereafter the increase in the value of f_v is only 20%. This occurs due to the fact that soot produced at the flame bottom, in most part, becomes oxidized or in other words is consumed at the top of the flame.

The observed behavior can be understood considering that the high exhaust gases temperature from the combustion causes the carbon molecules coagulation. This phenomenon leads to the formation of soot precursors (PAH), which then pass through the nucleation process, thus forming the particulate material. The highest f_v, 0.172 ppm, is observed for the equivalence ration $\phi = 8$, which was obtained in greater height measurement of 20 mm.

4.2 Comparison of the results with literature

The results of f_v obtained in this work were compared with related studies in the literature, which performed experimental studies and/or numerical modeling. These results

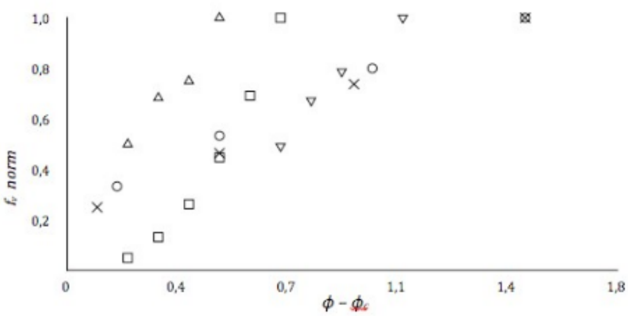

Figure 4: Normalized f_v values in function of the equivalence ratio ($\phi - \phi_c$) for the present study (x), in comparison to the results of [12], (△), [17] (□), [19] (▽) and [18] (o).

were normalized to allow comparison due to the fact that no publications with the same conditions and/or fuel used in this work.

Furthermore, as discussed in the previous section, there is a critical value for the mixture equivalence ratio, ϕ_c. Only for higher equivalence ratios, the formation of soot begins to be observed. This critical value was used to express the equivalence ratio of different works in absolute terms.

Figure 4 presents the comparison of f_v normalized values (maximum f_v value divided by the f_v value for each ϕ) with respect to the equivalence ratio in absolute values ($\phi - \phi_c$), always used the maximum values of f_v measured whatever the flame height. A local study was also used as basis of comparison, which developed a model of soot formation in a perfect stirred reactor, using n-heptane as fuel [19].

The behavior is similar for all the results from the literature analyzed, where the soot volume fraction increases as there is an increase in the equivalence ratio.

The increase in f_v occurred in a more sharply way in the experimental results obtained by Melton *et al.* [17], D'Anna *et al.* [12] and Hadef *et al.* [18]. The last one used ethylene as fuel and the others two, methane. This behavior is due to the flames configuration. These studies have used other types of burners, which is capable of producing flames of high power from high fuel rates. These characteristics produce flames concentrated in a small volume, reaching high temperatures, which favors the soot formation. On the other hand, the burner available in this study generates low power flames, which are important for model validations. Thus, there are no favorable conditions for the formation of soot that requires temperatures about 1,900 to be produced from methane [1].

The results of f_v in this work were compared to the theoretical results [19]. A very similar behavior was observed, despite the different conditions in which they were obtained. The correlation coefficient between the results is 99%.

The fitting curve containing the behavior of the results measured in this work may be described by the following equation, with the fit coefficient of $R^2 = 0.95$,

$$f_v = 0.26e^{0.98(\phi - \phi_c)}. \quad (2)$$

This equation can be used to calculate intermediate values or points beyond the measurement range used in order works to serve as base for future studies that aim to simulate the soot production in the same burner configuration used in this experiment.

5 Conclusions

The light extinction technique is a feasible way to measure the soot volume fraction produced in premixed flames, specially the flat flames which has a steady distribution of f_v in the transversal axis. There was no soot formation in the measurements made at the flame bottom, due to the quasi-stoichiometric combustion character in this region.

The values of f_v increased substantially in measurements made in heights above of 15 mm upstream to the burner surface. This behavior is consistent with the mechanisms of soot formation. For values of h higher than 15 mm, the growth of f_v tends to stabilize, due to the oxidation of the particulate matter produced.

The results found on the relationship between f_v and the equivalence ratio showed that the amount of soot increases exponentially with the equivalent ratio. With respect to mixtures of equivalency ratio greater than 4, excess fuel not burned is released in the exhaust gases, justifying the tendency of stagnation in the production of particulate material in very rich mixtures.

The use of the laser light extinction technique to determine the soot volume fraction led to experimental results similar to the ones obtained by the literature, when compared with each other through normalized values. Comparing to the theoretical model developed in a theoretical, the trend of increase of f_v with respect to mixture composition used showed consistent results. The correlation between the results of the two studies lies at 99%.

Also, was proposed a representative equation for the soot formation behavior as a function of equivalence ratio, which showed a correlation factor of 0.95. Future studies that require intermediate values in the f_v range, or even beyond this range, may use this relation, considering the uncertainty of 5%.

Measurements of f_v in this work may provide the basis for further studies in combustion, such as the numerical models validation of soot formation in premixed laminar methane flames, with equivalence ratio ranging 1.5 up to 8.

Furthermore, numerical simulations in perfect stirred reactor using natural gas as fuel can benefit from the results of this work to validate models.

Nomenclature

I_0 total intensity of light emitted by a laser, [mW]
I amount of transmitted light, [mW]
x flame thickness that the laser light crosses, [m]
f_v soot volume fraction produced, [ppm]
k_e light extinction coefficient, [-]
$\acute{V}_{air}$ volumetric flow of air, [m^3/s]
$\acute{V}_{fuel}$ volumetric flow of fuel, [m^3/s]
U_{global} global velocity of gases, [m/s]
R_e Reynolds number, [-]
h Flame height, [m]
u Uncertainty [-]

Greek letters

λ laser wavelength, [nm]
ϕ equivalence ratio, [-]
ϕ_c critical equivalence ratio, [-]

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