

Use of Spontaneous Chemiluminescence to determine the laminar burning velocity of syngas/methane mixtures

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Abstract In the present work we evaluated the use of CH* chemiluminescence to determine the position of the flame front as well as the laminar burning velocity (S_L) of some syngas-methane flames using the angle method that consist in calculate the angle of a Bunsen-type flame generated by a contoured slot-type nozzle burner. The syngas used in this study has a composition of 40% CO, 40%H₂ and 20% CO₂ (By Volume) and the addition of syngas to the fuel mixture (syngas-methane) was of 50%, 70% and 90% in volume, the measurements were carried out at 297 K and 1007.5 mbar of atmospheric pressure. An ICCD camera equipped with a colored filter centered at 430 nm was used to capture the chemiluminescence emitted by CH* and the digital photos obtained were stored as a pixel array of 1024×1024. Each pixel received a numerical value according to the intensity captured by the camera. The entire array was scanned then to locate the flame front, which is obtained in the pixels where the highest changes in intensity take place. A MATLAB code was used for this procedure. Numerical calculations of S_L were conducted using the one dimensional premixed flame code PREMIX of the CHEMKIN PRO package with the detailed kinetic mechanism GRI-Mech 3.0 in order to compare with experimental results. A good agreement between numerical and experimental results was obtained, which means that the determination of the flame front using CH* chemiluminescence in syngas-methane flames mixtures is adequate.

Keywords: Chemiluminescence, ICCD, image processing, laminar burning velocity, syngas, methane

1 Introduction

The understanding of phenomena involved in combustion processes is essential for a world who depends on such process to convert chemical energy in thermal energy. Through the years, it has been demonstrated that without an adequate knowledge of combustion parameters negative effects, like low efficiencies, high pollutant emissions and unstable operation conditions may prevail. Nowadays, all over the world the measurement of variables like velocity, temperature and species concentration are highly used to understand phenomena like heat and mass transfer which are really important in combustion processes. Generally, these measurements are taken using intrusive techniques that strongly affect the investigated combustion-based application being investigated and generate measurement errors [1].

Different optical diagnostic techniques have been implemented since three decades ago with satisfactory results in fields like reactive flows to determine combustion parameters in a reliable and accurate way. The most important characteristic of these techniques is that they are non-intrusive techniques and allow the monitoring of variables in real time during the experiments realization which has been constituted as a great advance in the study of heat and mass transfer phenomena [1], [2].

On the other hand, there is a worldwide interest in developing alternative fuels, which is of great importance for countries and regions with reserves of other alternative energy sources. Synthetic gas (syngas, SG) obtained from the gasification of coal and biomass is considered to be one of the most promising alternative fuels in developed and developing countries [3], [4], [5]. However, depending on the type of reactor and the gasifying agent, syngas generally has lower heating values between 1.0 and ~2.6 kWh/m³ and Wobbe index values between 1.5 and ~4 kWh/m³, which are very low compared to the values for pure CH₄ (9.425 kWh/m³ and 14.09 kWh/m³, respectively). Thus, there is a strong global effort to burn mixtures of conventional fuels and syngas, which also provides alternatives to increase the use of available fuels, but nevertheless, there is low information regarding to the combustion properties of this kind of fuels [6].

Laminar burning velocity (S_L) is one of the most important fuels and fuel mixtures property since it is

essential for characterizing several combustion processes. Information on S_L is fundamental for the analysis of the combustion phenomena such as the structure and stability of premixed flames, flashback, blow-off and extinction; turbulent premixed combustion; and the validation of reaction mechanisms in the presence of diffusive transport at high temperatures [7], [8], [9].

Due to the importance of S_L , different methods have been used to its determination. One of the most important, because of its simplicity and accurately, is the angle method that consists in calculate the angle of a Bunsen-type flame. However, to obtain a correct value of this angle it is necessary to determine appropriately the flame front by means of an optical technique. Determination of laminar burning velocity is possible by several optical techniques, for example taking direct photography of the luminous flame, taking schlieren or shadow photography, by means of chemiluminescence or by means of laser based techniques as Planar Laser-induced Fluorescence and Laser Doppler Anemometry [10], [11], [12]. In this work, the optical diagnostic technique implemented is based in chemiluminescence. Such process consists in the spontaneous emission of electromagnetic radiation due to chemical reactions in a spectral range between Ultra Violet and visual [2]. In the case of hydrocarbons like methane the peak of this emission is produced by CH and OH radicals [13], [14], [15]. The chemiluminescence emitted from these radicals can be used to register the flame front and determine the laminar burning velocity using the angle method.

In the present study, laminar burning velocities of syngas/methane flames were determined experimentally and numerically within a wide range of equivalence ratios (0.8-1.4) capturing spontaneous chemiluminescence (CH*). Syngas used in this study has a composition of 40% CO, 40%H₂ and 20% CO₂ (By Volume) and the addition of syngas to the fuel mixture was of 50%, 70% and 90% in volume, the measurements were carried out at 297 K and 1007.5 mbar of atmospheric pressure. The Experiments were conducted using an ICCD camera equipped with a colored filter centered at 430 nm, values of S_L were determined with the angle method and were compared with simulations performed with CHEMKIN PRO package using the reaction mechanism GRI-Mech 3.0.

2 Methodology

2.1 Experimental setup

Figure 1 is a schematic diagram of the experimental configuration implemented in this study. The flames were generated in three contoured slot burners with different outlet geometries. The selection of the burner depends on the estimated burning velocity of the mixture, considering that the slot output speed is directly related to S_L . The contoured slot-type nozzles (13.8mm x 5mm, 21mm x 6.7mm and 29.8mm x 9.4mm) helps to reduce the effect of flame stretch and curvature in the direction of the burner axis. A contoured slot-type nozzle also allows to have laminar Reynolds numbers for all the equivalence ratios to be studied, as well as nearly uniform exit velocity profiles. Additionally, a cooling water circuit inside the burner keeps the mixtures at a constant temperature.

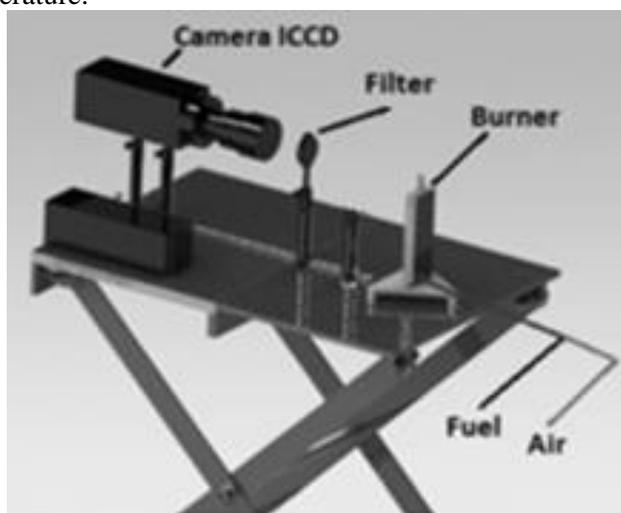


Fig 1. Schematic diagram of the experimental setup

Several lean and rich methane and syngas flames were generated in the burner described above. Table 1 lists the volumetric composition of these gases. The air was supplied by an air compressor and dried using two inline water traps. Each air-to-fuel ratio and exit velocity were ensured using rotameters that were specifically calibrated for each component gas, similar to those used in [7], [9]. The errors in the final composition were estimated to be lower than 2%. The syngas used in this study has a composition of 40% CO, 40% H₂ and 20% CO₂ (By Volume) and the addition of syngas to the fuel mixture was of 50%, 70% and 90% in volume, the measurements were carried out at 297 K and 1007.5 mbar of atmospheric pressure,

Table 1. Volumetric composition of syngas/methane used in the study

Mixture	Fuel composition
100-0	40% H ₂ + 40% CO + 20% CO ₂
90-10	36% H ₂ + 36% CO + 18% CO ₂ + 10% CH ₄
70-30	28% H ₂ + 28% CO + 14% CO ₂ + 30% CH ₄
50-50	20% H ₂ + 20% CO + 10% CO ₂ + 50% CH ₄
0-100	100% CH ₄

To determine the laminar burning velocity the angle method was used. The measurement is based on the principle that the velocity at the nozzle exit of the unburnt gases is equal to the velocity at which the flame front propagates from the burnt to the unburnt zone at an angle θ as shown in Figure 2. The laminar burning velocity is related to θ according to equation 1.

$$S_L = U \sin(\theta) \quad (1)$$

Where U is the mean velocity of the unburnt gases at the exit of the burner.

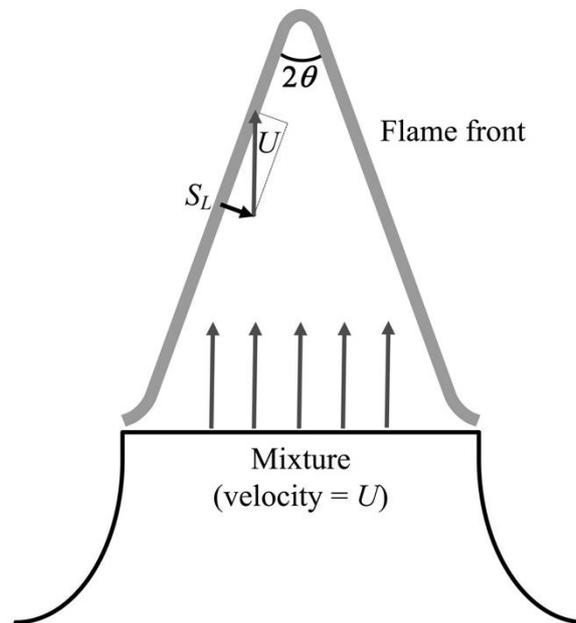


Fig 2. Burner stabilized flame technique (angle method)

The mean velocity at the exit of the burner nozzle was calculated from the nozzle area and the flows of the fuel-air mixtures while the flame angle was measured from chemiluminescence (CH*) photographs as follows. First, the digital photographs obtained by the ICCD camera are stored as a pixel array of 1024×1024. Then using a Matlab code a background image previously taken before experiments is subtracted and the flame is located where the maximum intensity is registered. The code detected the edges of the flame fronts, and thus the corresponding flame angles are calculated.

For the measurement of chemiluminescence, an ICCD camera (PI-MAX; Princeton Instrument) was used. To capture the light emission of the CH radicals (CH*), the lens was equipped with an interference filter. The center wavelength of the filter was 430 nm. The full-width-half-maximum (FWHM) was 11.02 nm and the minimum transmissivity was 45%. In chemiluminescence measurements, the CH* images were taken 25 times at each condition and the signal-to-noise ratio was less than 10% of the maximum intensity.

2.2 Numerical methodology

Numerical calculations of S_L were conducted using the one-dimensional premixed flame code PREMIX of the CHEMKINPRO package. The present simulations considered the GRI-mech 3.0 mechanism [16]. For an accurate calculation of S_L recommendations of Bongers and De Goey [17] were followed; transport properties were evaluated using the multicomponent diffusion model and thermal diffusion (Soret effect) was included in the calculations due to its importance on the hydrogen oxidation. Additionally, it has been reported that the accuracy of the calculated S is highly sensitive to the number of grid points used in the calculations; using a low number of points can lead to errors from 5 to 10% [18]. Therefore, according to Dlugogorski et al. [19], GRAD and CURV values were set lower than 0.01 to generate a grid of more than 1000 points, where S_L values converged and the flame temperature approached the adiabatic flame temperature. This same methodology has been used successfully to study other fuel gases [7], [9].

3 Results

Figure 3 shows an example of flame profile obtained in the experiment for methane without addition of syngas at equivalence ratio of 1.0. The reaction zone is clearly visualized by means of an ICCD camera equipped with a colored filter centered at 430 nm and the post-processing technique is able to determine the angle of this profile to compute the flame velocity of this investigation.



Fig 3. Instantaneous photography of flame front of methane at equivalence ratio $\phi=1.0$

The numerical and experimental results of the present work are shown in figure 4. In x direction is located equivalence ratios from 0.8 to 1.4, and in y direction the laminar burning velocity in cm/s. As syngas contains hydrogen and the laminar burning velocity of this is very high, S_L is higher for mixtures with higher content of syngas. This increase can be explained by the addition of H₂ to the mixture due H₂ promotes formation of OH radicals increasing the concentration of H producing an increment of the reactivity of the mixture and consequently of laminar burning velocity [20], [21], [22], [23]. At the same time the addition of syngas generates that the peak of S_L move to rich mixtures.

According with measurements the percent of increase in S_L peak from the mixture with 0% syngas to 50% syngas mixture is about 22 % which is in concordance with previous studies [20], [24]. And for this study S_L peak for the mixture with 90% syngas in content is two times S_L peak of pure methane.

Regarding with numerical results figure 4 shows good agreement between numerical and experimental data at lean conditions, however, GRI-Mech 3.0 underestimates S_L values for rich conditions especially near the maximum S_L value. This behavior has been already registered by Natarajan [25]. In this work, for peak S_L , the maximum error between numerical and experimental results is 12%.

With the increase of S_L , for the syngas/methane mixture, the blow off tendency is expected to improve when compared to a flame of pure methane.

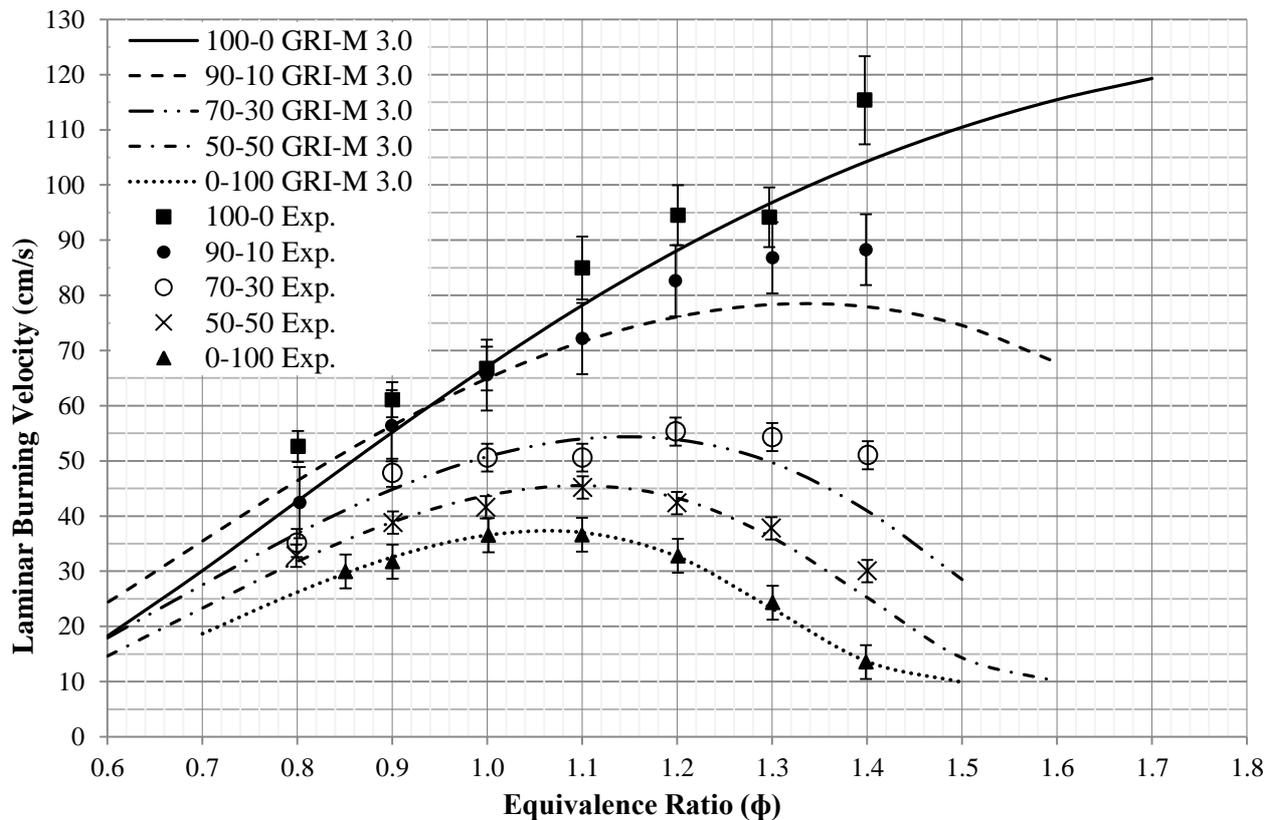


Fig 4. Experimental and numerical results for laminar burning velocities at 1007.5 mbar

4 Conclusions

Measurements of the laminar burning velocity of syngas/methane mixtures were made using chemiluminescence of CH radicals. Numerical calculations of S_L using GRI-Mech 3.0 were also performed to be compared with experimental results and a good agreement was found besides measurements agree with reported literature. Therefore it is possible to conclude that use of chemiluminescence to determine S_L is adequate.

With this study it is concluded that diagnostic optical techniques and in particular chemiluminescence are strongly important for combustion analysis because allow monitoring the flame in non-intrusive way.

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