Evaluation of chemiluminescence as sensor for lean premixed combustion

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The purpose of this research is to establish the validity of using chemiluminescence as sensor in an active control combustion loop. The main objective is to evaluate which are the required conditions in terms of equivalence ratio, integration time and fluid motion to get a reliable equivalence ratio measurement using chemiluminescence emission. Measurements are performed in two different kind of combustor: a swirl type combustor (15kW) and a pilot flame burner (90kW). It is shown that equivalence ratio down to 0.40 can be measured using ratio of OH^{*} versus CH^{*} which shows good sensitivity to equivalence ratio. As high acquisition rate can be achieved using an AD converter, the frequency domain is investigated. One can notice that the oscillations of the flame can be detected using chemiluminescent emission and that those frequencies are similar to the one obtained with a pressure probe. Finally, wavelet transform is applied to time series chemiluminescent signal to detect the appearance of the different oscillating modes present in the demonstration combustor. It is shown that two modes are alternatively the main mode.

1 Introduction

Efforts are being made in order to reduce pollutant emissions (especially NOx) from gas turbine. In order to achieve such reduction, an attractive issue is the lean premixed combustion. This type of process however, is known to lead to instabilities, leading to possible blowout. For safety reasons, it is common to run the combustor with a wide margin above the lean blowout limit. In order to limit further the emission, the operating point should be closer to this limit and therefore a precise monitoring of the conditions and possible control strategies to avoid blowout have to be included while operating the combustor. When dealing with active control strategies, the definition of the proper quantities to measure is very important. For combustion case, the harsh environment increases the difficulties and the detector has to sustain high temperature and preferably be non-intrusive. The monitored quantities should depend on the causes of instabilities. Those can either result in a change in equivalence ratio (ER) ([1]) or in thermo-acoustic interactions [2] and therefore quantities sensitive to stoichiometry and acoustics should be simultaneously monitored. Acoustics properties can be measured with pressure probes, whereas stoichiometry quantities are more difficult to access. In fundamental studies, it is possible to use ion-probe current ([3]) but its application to practical setup is quite difficult. An alternative way would be to use diode laser absorption techniques. Very good results in using them for active control have been already reported ([4]), however, their application to practical environment may also be difficult. Therefore, chemiluminescence measurements are attractive, as they do not require any actuator to be activated and only an optical access may be required. The drawback is that it is therefore very difficult to precisely know what is measured and the optical properties of the glass may change with time. One possible way is to take not the intensity of a wavelength, but the ratio of the intensity for two different wavelengths. This procedure enables to minimize the secondary effects as may come from detecting devices, optical access. The use of intensity ratio is not new ([5, 6]), but the presented results are mainly for flames more rich than 0.50. The main question is towards which limits those measurements are reliable. The aim of the present investigation is to evaluate both the leanest limits of ER measured as well as typical temporal and spatial consideration for its application. The first part will present the principle of the technique, relying on chemiluminescence. The second part will deal with experimental validation of the technique for various stoichiometries under different dynamics conditions (velocity effects are considered) in a simple swirl combustor. Finally, applications in a demonstration combustor are reported, for which instabilities occur. Those instabilities are also treated in the frequency domain.

2 Experimental setup and preliminary results

2.1 Formation of chemiluminescence

The formation of chemiluminescent products in combustion is still a challenging issue for modeling all the kinetics of reaction. Among the main products in methane-air flames, OH^* , CH^* can be detected. As the present flames are towards lean limits of stoichiometry, the appearance of C_2^* in excited states is too weak to be considered. On the other hand, as this control strategy should be able to deal with real conditions (high pressure), one has to consider the CO_2^* broadband emission. The formation and evolution of OH^* , CH^* and CO_2^* may be described by the processes described in Table 1.

$$CH + O_2 = CO + OH^* \qquad C_2H + O = CO + CH^* \qquad CO + O + M = CO_2^* + M$$
$$C_2H + O_2 = CO_2 + CH^*$$

Table 1 Possible formation of excited molecules

From excited state to normal state, two possible mechanisms exist. Spontaneous emission of photon through a reaction $D^* \rightarrow D+hv$ (where the energy of the photon is a signature of the molecule as it is function of electronic states of the excited molecules) is the chemiluminescent emission. Another process of de-excitation is through collisional quenching where the mechanism is $D^{*+}M\rightarrow D+M$. Typical chemiluminescent emission are reported in Table 2. A remarkable molecule is the CO_2^* that is emitted not on precise wavelength but that is broadband and therefore as to be taken into account when computing emission level of the other species.

Radical	Transition	λ (nm)
OH^*	$A^2\Sigma^+ \rightarrow X^2\Pi(\Delta\nu=1)$	282.9
OH^*	$A^2\Sigma^+ \rightarrow X^2\Pi(\Delta \nu = 0)$	308.9
CH^{*}	$B^2\Sigma^- \rightarrow X^2\Pi$	387.1
CH^*	$A^2 \Lambda \rightarrow X^2 \Pi$	431.4
CO_2^*	Continuum	300→500

Table 2 De-excitation process from chemiluminescent emission

Typical constant rates are still investigated for those processes, as recently reported ([7-8]) and comparison between predicted emission and measured emission are qualitatively similar.

2.2 Experimental setup

To test the relations existing between chemiluminescence ratio and stoichiometry, a swirl-stabilized flame is used. The swirl base has a diameter of 38mm and the inner diameter is 16mm. The swirl is composed of 12 vanes each having a constant angle of 15° , which gives a swirl number of 0.23. Inlet velocity and mean ER are changed to measure their effects. A sketch of the facility as well as a picture of the injector may be seen in Figure 1. To constitute the combustor chamber, a VYCOR glass is used with a length of 20cm. The air is heated before entering the mixing chamber to have a value of 600K when entering the test section. The mass flow rate of both air and methane are monitored. A thermocouple is placed after the mixing chamber to measure the inlet temperature of the mixture. All quantities are continuously acquired via a PC and the data are afterwards treated to evaluate the relative differences in inlet conditions. The results are that typical fluctuations of ER are lower than 0.01, the uncertainties in velocity are of the order of 0.2 m.s^{-1} whereas temperature fluctuations are within 2K.



Figure 1 Sketch of the swirl-stabilized flame experiments

2.3 Evolution of flame as function of ER

The influence of stoichiometry is first measured with natural emission and OH^{*} chemiluminescent images. The camera used is an ICCD camera, with a spatial resolution of 1024 by 1024 and a dynamic range of 16 bits. To get the maximum resolution, the gain as well as the shutter time is changed according to the imposed stoichiometry. The field of view is set to 70 by 70 mm, with bottom view at the exit of the swirl. The purpose is to evaluate the measurement points of photo multipliers linked to the filters. Both natural emissions (obtained with no filter and a digital camera) as well as only chemiluminescent are shown here. As can be seen in Figure 2, a decrease of ER leads to a lifted flame and a necking of the flame. This finally will lead to blow off. The images show that the

flame is symmetric and that the point of maximum emission is lifted with a decrease of ϕ . The integration time has to be increased to get a good signal from 1ms for an ER of 0.80 to 10 ms for 0.47 (the exit power drops from 24 to 14 kW). An ER of 0.47 is the lean blowout point for a velocity of 15 m.s⁻¹. For lower velocities, a lower stoichiometry can be achieved and hence higher lifted height can be reached. However, the lowest velocity investigated here is 12.5 m.s⁻¹, for which the lifted height did not exceed the 20mm, location of the measurements. The red line shown in all figure is the line of measurements performed hereafter with photo multipliers. This line always corresponds to a combustion zone that is the most important condition to fulfill. It can also be noted from the natural emissions, that the flame is a blue flame, and therefore no influence of black body emission is expected.



Figure 2 Chemiluminescent measurements as function of stoichiometry

3 Measuring equivalence ratio with chemiluminescence

3.1 Experimental setup

An important issue to address is the type of collecting probe that should be used for sensing ER. A too broad sensor may introduce bias by taking into account other species than those created typically at the flame front, which is the purpose of the present diagnostics. On the other hand, a too narrow system may give local indication, but with the major risk of missing the general instabilities. Therefore, the present sensor is based upon the measurement with a lens system (focal length of 30cm), allowing typically all the light issued in a cone to be sampled. This light is hereafter sent to photo multipliers (PM) through a series of dichroic mirrors (DM) and band-pass filters (BPF). The intensity measured is proportional to the number density of excited molecules within the probe volume. The typical optical arrangement is displayed in Figure 3.



Figure 3 Sketch of the photo multipliers arrangement

An important characteristic is that the light is split by each dichroic mirror, resulting in a decrease of initial

intensity for each wavelength. However, each dichroic mirror is acting as a band-pass filter reflecting mainly the light of the specific wavelength and transmitting the other wavelength with a ratio of 90 to 95%. The emission around 473nm is usually used to monitor C_2^* chemiluminescence. However, it has been shown that for lean combustion, C_2^* emission is negligible and that for this wavelength the main species emitting is the broadband CO2^{*}. The second wavelength of C_2^* is not used in the present study.

3.2 Chemiluminescence measurements for different ER and mass flow rate

The first task before trying to measure ER is the determination of the response of the sensor for different ER and inlet velocity. For this purpose, the lens is mounted on a traverse system and systematic measurements are performed to determine the variation of emission as function of stoichiometry, mass flow rate and position. The intensity is varying with the position of measurement also, but only measurements made for a height of 20mm are reported for this case. To see the evolution of chemiluminescence as only a function of stoichiometry, they are corrected with the initial flow rate. It is found in Figure 4 that the three different species can be approximated by some functions, which expressions are given in equation 1. The quantities in these equations are the emission corrected by the mass flow rate. Those functions are determined by being the one giving the highest regression coefficient for the species. The lines of Figure 4 are made upon a best-fit function, which expression is

$$OH^* = 8664\phi^{6.09}$$

 $CH^* = 0.223 \exp(9.89\phi)$ (1)
 $CO_2^* = 0.098 \exp(9.99\phi)$

As it can be seen, the evolution law of OH^* and CH^* , CO_2^* are different. The OH^* emission is a power function of stoichiometry, as reported by [9]. This difference explains also the non-monotonic behavior of the ratio made with OH^*/CH^* and the monotonic ratio of CH^*/CO_2^* that is an exponential function of ER. The present coefficients are valid only for this specific equipment and setup and any change in the collecting light probe and/or in the setup will result in other coefficients, but the functions should remain identical for stoichiometry lower than 0.80. For higher ER, it has been reported a decrease of chemiluminescent emission (see [10]). The point of higher ratio between OH^* and CH^* , depending in the coefficient will also vary with experimental conditions.



Figure 4 Emission for different flow rate and ER

From these emissions evolution, one can compute the ratios of different species. As emphasized by the nature of the approximation function, one expect the ratio of OH^* versus CH^* not to be monotonic, whereas the ratio of CH^* versus CO_2^* should be monotonic. This is illustrated in Figure 5 where one can see that there is a maximum for an ER of 0.65 in the case of the first ratio. The absolute value of the maximum should depend strongly on the configuration of the experimental setup, but the position of the peak should be quite stable, as being linked with chemistry. On the other hand, one can notice a decrease of the second aforementioned ratio with the stoichiometry. This has a direct consequence of giving a possibility to identify clearly the ER, having a ratio being quite sensitive to stoichiometry and a second, less sensitive, but presenting monotonic behavior.

To verify the present approximated function, the experimental OH^*/CH^* ratios are plotted together with the ratio derived from the interpolated functions. Results presented in Figure 6 show that good agreements are found for lean stoichiometry conditions. The theoretical curve is the one obtained with the interpolated functions. One can see that the ratio is indeed independent of the velocity for the lean cases. For higher ER, the influence of C2^{*}

emission is not negligible in the determination of CO_2^* emission and should be taken into account, which is not the case here. This explains the influence of velocity in the rich side of the graph.



Figure 6 Comparison between the approximated function and the experimental results

3.3 Temporal considerations

An important feature of the present technique is its temporal behavior to get reliable results. It is important to understand the typical integration time required to obtain a good measurement. For this purpose, the signal is acquired for 5 seconds at a sampling rate of 10kHz. The signal is hereafter analyzed by segments of identical duration. The ER is computed according to the previous ratio of functions. The results reported in Figure 7 are obtained with the signal taken from the origin of time to the measurement time Δt to evaluate the typical time required to get stable results. This assumes no local fluctuations in the ER. This assumption is made as no oscillating combustion was detected for this combustor. As it can be seen, a stable value is achieved after 0.3s and remains afterwards constant. The variations at the initial stages are very important as they show systematic under-estimation of the ER. Therefore evaluating whether there is always an underestimation of the ratio or if this is part of uncertainties estimation is important.



Figure 7 Temporal evolution of the ER measurements

To verify that the ratio may be sometimes underestimated and sometimes overestimated the signal is decomposed into several sub-signals with varying the temporal origin and each sub-signal is then individually processed to determine the ER by taking mean chemiluminescent emission within its duration. The results obtained are displayed in Figure 8. The acquisition time represents the duration of the sub-signal considered and the estimated ER is obtained by taking the average of chemiluminescence signals within this time duration minus

the background level as determined without flame. The line represents the average over all sub-signals for the acquisition time chosen and to evaluate relative error, the mean value is set to true ER; therefore we assume no systematic error in measuring ER. The lower and higher estimation respectively represents the lowest and highest ratio measured obtained within the 450 sub-signals. One can notice that there is no systematic under-estimation and that the extremes tend to be constant after 0.3s. This means that no benefit is obtained by considering a bigger integration time. The typical uncertainty is estimated, according to Figure 8 to be 5% but it is increasing with a decrease of ER and drops to about 10% for stoichiometry of 0.40.



Figure 8 Temporal evolution of ER with varying temporal origin

4 Application to demonstration combustor

4.1 Presentation of the setup

To assess the usefulness of this technique, measurements are performed in the demonstration combustor of the project. This combustor has a premixed pilot burner and a main mixture, which is injected through a conical flame holder. It is composed of 8 slits, which begin 285mm from the exit of the combustor, and end 225.5mm from this exit. The combustor has a squared cross-section of 100mm and a length of 213.5mm. The pilot burner exit is located 89.5mm upstream the end of the conical flame holder. The distance between the main mixer exit and the combustor is 45.8cm. Typical output power is 90kW for a velocity of 15m.s⁻¹ and ER of 0.50. A sketch of the combustor is shown in Figure 9. The pressure transducer (model XTME-190-25G) is placed 17.9 cm from the exit in the middle plane. Its sensitivity is 2.617mV/PSIG. Chemiluminescent measurements are performed in a plane parallel to the main flow direction. Those measurements serve at determining the points for active control monitoring, which should be independent from pilot percentage and be a function only of ER. Placing the measurement point just after the exit will make it mainly sensitive to the pilot flame, whereas putting it too far will lead to measure mainly hot gas emission and therefore not be directly a function of ER. Another important point is the direction of measurements.



Figure 9 Sketch of the demonstration combustor and actual picture

The lens (focal length of 30cm) monitoring system is placed perpendicularly to minimize the influence of black body emission that arises from the flame holder structure. Furthermore, if the lens is not placed perpendicularly,

it will integrate the light from different regions of the flame and maybe may not be directly sensitive to ER. The stoichiometry of the pilot is different than the one of the main mixture and the correspondence is given in Table 3. The ER corresponds to stoichiometry value of the overall mixture and the values in the table are the one for the pilot. For the lowest case of pilot percentage, the values presented here are estimated using linear regression as no direct measurements have been performed. For higher percentage (2.00 and 3.00%), the values result from direct tests.

Pilot	ER 0.44	ER 0.50	ER 0.55	ER 0.65	ER 0.70
1.20%		0.35	0.39	0.46	0.49
2.00%		0.55	0.60	0.71	0.76
3.00%	0.75	0.85	0.94	1.10	1.19

Table 3 Pilot ER as a function of main ER

4.2 Chemiluminescent measurements and ratio determination

Chemiluminescent measurements are performed for different conditions of pilot burner percentage and global ER. The main problem is the attenuation of the light due to a deposition on the glass for a global ER higher than 0.55. This behavior is shown in Figure 10 where the OH* intensity is plotted for different pilot percentage and global ER. The location of the measurements is 180cm from the exit of the combustor, which is the position of the pressure transducer. Therefore, for these conditions, it is impossible to approximate the different chemiluminescent emission as a function of ER. However, as the changes in transmissivity of the glass should affect all the wavelengths, it is expected that the chemiluminescent ratios be not affected by these changes. To measure the chemiluminescent ratio of OH* versus CH* and CH* versus CO2*, first the spatial distribution of chemiluminescence is measured. Based upon the results obtained, the measuring point of intensity ratio is decided to be in the centerline and at a distance of 180mm from the exit of the combustor, which corresponds to the location of the pressure transducer. The reason of this choice is that it is the point showing the least influence of the pilot flame on the ratio being affected only by global ER (see Figure 11). Therefore the influence of ER on this ratio is shown only for this specific point. The influence of ER is reported in Figure 12 for the ratio of both OH* versus CH* and CO₂* versus CH*. As it can be seen the ratio CO2* versus CH* is monotonic, as previously reported for the simple swirl combustor. The exponential decrease of the second ratio (that was predicted by the first measurements in the swirl type combustor) is also seen for this kind of geometrical configuration. This second ratio does not suffer from any influence of the pilot percentage that is less the case for the OH^{*} emission.



Figure 10 OH^{*} chemiluminescent measurement as a function of global ER and pilot percentage



Figure 11 Influence of pilot on intensity ratio as function of stream wise position of measurements



Figure 12 Influence of stoichiometry on intensity ratio

4.3 Frequency content

An important feature is evaluation of oscillatory combustion that is a key point in active control combustion strategy. To evaluate the possibility to use chemiluminescent emission as monitoring instability, simultaneous measurements of pressure and OH^{*} are performed for different conditions of inlet velocity and pilot percentage. The acquisitions were performed simultaneously at a rate of 10kHz for the four channels (one pressure and three chemiluminescent), allowing spectrum up to 4kHz to be computed. Usually, only pressure signal spectral content is used [11-12] to determine instabilities frequencies. As depicted in Figure 13, the peak detected in the chemiluminescent signal and the one issued from pressure measurements match perfectly. This means that the signals can be alternatively used as oscillations monitoring device, the chemiluminescent measurements having the advantage of giving also the ER. Therefore, afterwards, only spectrum obtained with chemiluminescence is shown, knowing that the frequency obtained is also measured in pressure signals. The measurements show a frequency of 496Hz and its sub-harmonics in all cases. This corresponds to an acoustic wave issued from the main mixer, according to quarter wavelength rule. The influence of the pilot flame on the stability of the combustion can be seen in Figure 14 where the cases of three different oscillating conditions are displayed, with only an increase in pilot percentage as changes between the tests. The overall ER for those measurements is 0.70 and the pilot is changed from 1.20% to 3.00%. The increase of pilot percentage results in an appearance of a secondary oscillating mode, which is not the primary acoustic instability. This later amplitude decreases to become less important than the secondary peak for pilot percentage of 3.00%. This secondary peak corresponds to the quarter wavelength rule applied to the distance between exit of the pilot to exit of the combustor, therefore it is increasing with an increase of pilot percentage. Its effect is also to reduce the amplitude of the primary frequency by splitting the energy into two oscillating modes, which can be very useful if this primary instability becomes large enough to cause damages. This may be used as active control when the primary instability becomes large.







Figure 14 Influence of pilot percentage on oscillating combustion

However, increasing pilot leads also to an increase of NOx and therefore optimum point is to be decided.

Table 4 shows the difference overall ER as well as pilot flame percentage used and the cases where oscillator	ry
combustion is detected (peak of frequency or frequencies) are explicated with their frequency, whereas cases n	ot
leading to oscillations are marked with No.	

Pilot percentage	E. R. 0.44	0.50	0.55	0.65	0.70
1.2 %			No		496
2.0 %			No	480, 780	496, 810
3.0 %	No	No		480, 780	496, 810

Table 4 Experimental conditions and oscillatory combustion cases

As it can be noticed, oscillations are mainly present for ER higher than 0.55. The relative importance of the main acoustic mode and the pilot flame oscillation is varying with pilot percentage. The difference in the excited frequencies between the different ER comes from a change in the mean temperature of the flame with the ER. Typical flame temperature is 2000K for 0.70 and only 1930K for 0.65.

4.4 Wavelet analysis

As seen in Figure 13, two modes are present, but no details concerning temporal signature of each one can be found by Fourier transform. In order to better characterize the modes of instability, wavelet transforms are used on the OH^{*} chemiluminescence signal with a global ER of 0.70. For a detailed explanation of wavelet transform principles and analyses, see [13]. In the present case, the second derivative of Gaussian, up to a constant, also known as "Mexican hat" is used. This wavelet base is similar to the one used in [14], but it is presently applied to chemiluminescence and not acoustic signals. Performing the wavelet transformation of known sinusoidal signals gives correspondence between scales and frequencies, for this specific acquisition frequency. The frequency is found to be is inversely proportional to the wavelet scale. Computing the continuous wavelet transform onto the basis formed by scales representing frequencies from 50 to 1000 Hz with a step of 2.5Hz enables to attribute to each point a specific scale (hence frequency). The selection of the proper scale at a given time is done on the basis of the coefficients for each scale, the highest giving the presently most energetic frequency. The results for two segments of signal are reported in Figure 15, where the y-axis corresponds to the instability (1 for the one of 496Hz; 2 for the 810Hz). Two different time sequences are chosen here to show that typical patterns produced. The symbols have been artificially moved to allow a better distinction between each. As it can be seen, the case with a pilot of 1.20% is mainly driven by the lowest frequency, and seldom by the higher mode. With an increase of pilot, the number of events attributed to the second instability increases.



Figure 15 Switching instability modes as detected by wavelet transform

For the lowest pilot percentage, one can notice from time to time a complete switch to the first instability (496Hz). The appearance of the secondary instability is noticed by the progressive switching to this higher mode and then again a decrease to only one oscillating mode. This secondary instability is linked with the pilot acoustic. However, no constant time-interval between those events is presently detected in the signals.

5 Conclusions

Systematic chemiluminescence measurements have been performed in two different kinds of combustors, one being a simple swirl type, the other being more sophisticated and closer to real applications. Those measurements consist in monitoring three different wavelengths, one corresponding to OH^* emission, one to CH^* and the other one mainly from CO_2^* broadband emission. The ratio of the intensity of those species can give the equivalence ratio (within 10% for the leanest cases investigated at the present time), as it is only a function of stoichiometry and not of quantities such as fluid motion. Even when the optical access' properties change with time, the ratio can still give reliable measurements. Typical acquisition frequency should be adjusted to respect Nyquist criteria for frequency analysis. Therefore, an interesting point is that its frequency analysis is similar to the one performed on pressure and therefore can be used as sensor for combustion instabilities in active control loop. Increasing pilot flame percentage leads to a lowering in the primary acoustic energy by distributing it to a secondary instability. Wavelet transform showed that both frequencies are alternatively excited but with different duration. For some conditions, one mode is very unstable and sometimes disappears for several milliseconds.

The next experiments will consist in linking the on-line measurements of ER and combustion instabilities to mass-flow controller in order to regulate the pilot percentage as well as the global ER for active control strategy. Further processing (on-line if possible) is required on time-series signal to detect as early as possible the blow out events. Further analysis and experiments will also be conducted on chemiluminescent ratios and effects of other parameters (like inlet temperature) will be done to extend the actual database and compare with OH-PLIF/PIV measurements to understand flame dynamics. The final stage being the application of the monitoring device to pressurized flames.

6 References

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