Possibility of applying flame chemiluminescence and ionization current to the combustion status monitoring

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1 Introduction

The European natural gas supply is expecting larger variations of gas composition in the future, due to a more diverse set of supply sources but also the integration of renewable energies (e.g. biogas and hydrogen). For gas boilers, the calibration with a given fuel will then be insufficient to ensure an optimized performance especially in terms of efficiency and pollutant gas emission. The development of an auto-adaptive combustion control system has thus received interests from gas boiler manufacturers. For this purpose, the first step is to gain reliable and precise information of the combustion status. One important parameter to monitor is the air-fuel equivalence ratio. Several solutions have already been proposed for it. In the present study we will be focused on two of them, i.e. flame chemiluminescence and flame ionization.

The flame chemiluminescence is easy to record and is especially sensitive to the equivalence ratio. The diagnostic technique is mainly based on the emission intensities of OH^{*} (308 nm), CH^{*} (430 nm), C^{*}₂ (516 nm) and the broadband CO^{*}₂ (between 300 and 600 nm, often represented by the value at 450 nm). Its application to real systems has recently been reviewed in [1], and the control of the equivalence ratio status has been proven in [2]. However, the feasibility study with different compositions of natural gas is incomplete.

Another potential solution concerns the flame ionization. The flame can be considered as a weakly charged plasma. When a voltage is applied, an ionization current can be measured. Its intensity is shown to be related to the flame equivalence ratio. Applications of the technique to household gas boilers are emerging [3,4]. Nevertheless, a more comprehensive investigation on the effects of various factors (power, gas composition, probe position, etc.) is still needed to evaluate its applicability to the flame diagnostics.

In the present study, a laboratory premixed conical flame is used to infer the effects of fuel variations on the equivalence ratio monitoring with the two techniques. The first part focuses on the flame chemiluminescence with different natural gas compositions. Apart from the main component of CH_4 , five major secondary

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components are considered: N_2 , CO_2 , H_2 , C_2H_6 , C_3H_8 . Their effects on the flame chemiluminescence of CH^{*}, OH^{*} and CO^{*}₂ are studied and the validity of the commonly used equivalence ratio indicator CH^{*}/OH^{*} is verified. In addition, direct numerical simulations with the REGATH code [5] are conducted to provide a support for the experimental results. The second part is dedicated to the flame ionization current. A series of tests are performed to understand the impact of flame power, supply voltage, probe position and fuel composition. At the end, the curves of ionization current vs. equivalence ratio are constructed with different fuel scenarios.

2 Test bench

The study is performed with a conical flame burner. Measurement facilities are shown in Figure 1. The chemiluminescence spectrum is measured with an OceanOptics USB2000+ spectrometer (200 - 900 nm). The scope is a circular area of about 100 mm diameter, which incorporates the longest flame encountered during the study. For the measurement of ionization current, a cylindrical probe (2 mm diameter) is installed horizontally above the burner exit. The position of probe tip is specified in the following by the coordinates (x, z) as shown in Figure 1 (right). A DC voltage is applied between the probe and the metallic burner exit, and the former serves always as anode.



Figure 1: Measurement of flame chemiluminescence (left) and ionization current (right)

3 Flame chemiluminescence

Secondary components are progressively added to methane from X = 0 to 20% where X represents the volumetric fraction, while the power and equivalence ratio are kept constant. The chemiluminescence intensities of OH^{*}, CH^{*} and CO₂^{*} are normalized by their values for the pure methane case, and drawn against the dilution fraction X in Figure 2 (circles with error bars). At the same time, a numerical calculation is conducted and the results are presented as dashed lines in Figure 2. The numerical results for N₂, CO₂ and H₂ dilution are based on the GRI-Mech 3.0 augmented with OH^{*} and CH^{*} related reactions [6]. Additional species and reactions are included for C₂H₆ and C₃H₈ [7].

The experimental measurements are first performed with a fixed power (P = 1.3 kW for N₂ and CO₂, P = 1.5 kW for H₂, C₂H₆, C₃H₈) and equivalence ratio ($\phi = 0.83$), while the volumetric fraction of secondary components varies from X = 0 to 20%. When the secondary component is also a combustible (H₂, C₂H₆, C₃H₈), the power and equivalence ratio values refer to the air/methane/secondary component mixture. It is shown that:

- The dilution of N₂ only slightly reduces the CH^{*}, OH^{*} and CO₂^{*} emissions (< 5% for $X(N_2) = 20\%$)
- The dilution of CO₂ causes a small decrease of CH*, OH* (around 10% for $X(CO_2) = 20\%$) and a slight increase of CO₂^{*} (< 5% for $X(CO_2) = 20\%$)
- H₂ barely changes the emission of OH^{*}, but causes a clear decrease of CH^{*} (> 15% for N₂ = 20%) and a small decrease of CO₂^{*} (< 10% for $X(H_2) = 20\%$)
- Addition of C_2H_6 or C_3H_8 causes a clear increase of OH^{*} and CO_2^* (> 10% for X = 20%), and a remarkable increase of CH^{*}(> 150% for X = 20%). For the same volumetric fraction, C_3H_8 has a stronger impact on the chemiluminescence intensities than C_2H_6 .

The most remarkable is the effect of C_2H_6 or C_3H_8 on CH^* as even a modest fraction causes a significant increase. One explanation is that the C_2H_6 and C_3H_8 , thanks to the longer chain of carbon, facilitate the formation of C_2H species, predecessor of CH^* .



Figure 2: Evolution of chemiluminescence intensity with secondary component fraction

The numerical results are generally correct in the prediction of qualitative trend and magnitude. However, a gap is noticeable between the experimental and numerical results, especially in the case of C_2H_6 and C_3H_8 . This may result from an unoptimized reaction mechanism and an over-simplified calculation of flame surface area. However, the optimization of the numerical model is not the main purpose of the present study.

The CH*/OH* ratio is now investigated as a function of equivalence ratio for pure methane and five binary mixtures consisting of methane and one secondary component each time. The volumetric fractions of secondary components are as follows: $X(N_2) = 10\%$, $X(C_2) = 10\%$, $X(H_2) = 20\%$, $X(C_2H_6) = 10\%$, $X(C_3H_8) = 5\%$. They are chosen to represent the maximum value seen in real natural gas (also considering the injection of biogas and hydrogen). Tests are performed at a unique flame power due to a limited range of stabilization, but the ratio of two chemiluminescence intensities removes a priori the impact of

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Figure 3: CH*/OH* vs. equivalence ratio under different fuel composition scenarios.

flame power. The resulting curves are shown in Figure 3. It is found that the calibration curve obtained with pure methane (black dashed-dot line) remains valid for moderate dilutions by N₂ or CO₂. Addition of H₂ increases a noticeable error of the predicted equivalence ratio ($\Delta \phi \sim 0.05$ for $X(H_2) = 20\%$). For C₂H₆ and C₃H₈, however, even a modest fraction of a few per cents in the fuel mixture causes considerable errors ($\Delta \phi > 0.1$), which may not be acceptable for combustion control. The gaps for the curves of H₂, C₂H₆ and C₃H₈ with regard to pure CH₄ result clearly from the different effects of these components on CH^{*} and OH^{*}.

4 Flame ionization current

The effects of probe position, power, voltage, equivalence ratio, and secondary fuel components on the flame ionization current intensity are now investigated.

The ionization current with different probe tip positions is measured and presented in Figure 4 (left). It is found that when the probe is in contact with, or very close to (less than 3 mm), the flame front, the current intensity barely changes with the probe position. It needs to be pointed out that the conical flame is largely deformed by the probe when the probe is in contact with the flame front, but this perturbation does not change the current intensity. If the probe is moved further away, a decrease of the current is indeed observed, until a final drop to zero when the probe is completely outside the zone of burnt gases.

Figure 4 (middle) shows the evolution of flame conductance (ionization current normalized by supply voltage) with gas velocity (flame power), for different probe positions and supply voltages. For z = 2 cm, the probe is always in contact with the flame. The ionization current is observed to decrease with the increase of gas velocity from 1.2 m/s. For z = 5 cm, the probe is in contact with the flame only at higher flowrates. We notice in this case an increase of the current intensity at the beginning and a drop for the second half. When the flowrate increases, the flame height increases as well and consequently the distance between the flame front and the ionization probe progressively reduces before the ionization probe protrudes within the flame front only at the highest injection gas velocities and an ionization current increases roughly monotonically. In addition, by comparing the solid and dashed lines, we find that a higher supply voltage tends to enhance the current intensity, especially at high flowrates close to blow off limit.

The effect of secondary fuel components is studied with the similar method described in section 3. The



Figure 4: Effect of probe position (left), flame power (middle) and secondary components (right)

evolution of flame conductance with dilution is shown in Figure 4 (right). We find that the dilution of N₂ and CO₂ causes a decrease while the other components cause an increase of ionization current, except that the C₃H₈ curves level off between X = 15% and 20%. Also, a higher supply voltage results in a higher flame conductance. The validity of ionization current as an equivalence ratio indicator is then tested, and results are presented in Figure 5. We remind that, since the tests above have shown a power-dependency of the ionization current, these curves are applicable only to one unique flame power (P = 1.5 kW). It is found that all the secondary components cause certain drifts from the pure methane curve but the gaps remain relatively small. The errors are therefore moderate ($\Delta \phi < 0.05$) for the mixture fraction exploited. However, bigger errors are expected with higher dilution fractions, for which biogas, containing up to 50% CO₂, is an example.



Figure 5: Ionization current vs. equivalence ratio under different fuel composition scenarios.

A complete theoretical explanation for the above findings on the ionization current is difficult at the current stage of the study. A qualitative description is intended here. According to Rodrigues *et al.* [8], the major drop of electric potential takes place at the cathode, which is also the decisive factor of ionization current intensity. This may explain why the position of ionization probe (anode) is unimportant as long as it is in contact with the flame front. We may also deduce that the flame stand-off distance (or dead space) is an important factor, since a smaller stand-off distance should help reduce the huge resistance near the burner rim (cathode) and facilitate a higher ionization current. Former studies have shown that the stand-off distance is decreased with a high supply voltage [9, 10], an equivalence ratio close to stoichiometry [10], or a low gas flowrate [11, 12]. These trends are quantitatively coherent with the evolution of ionization current.

Further investigations are still needed for a better understanding of the phenomena.

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