Influence of Hydrogen distribution on flame propagation

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1 Introduction

For environmental reason, hydrogen can be considered as a fuel of the future. However, the flammability domain is very large and the necessary ignition energy is only of few millijoules which makes the presence of hydrogen in air potentially dangerous in confined environment. It can also be accidentally generated in nuclear plants. The expanding flame is intrinsically unstable. Numerous studies, for example Lee et al. [1], Knystautas et al. [2], showed that obstacles located along the path of expanding flames can cause rapid flame acceleration and deflagration to detonation transition. Generally, if the hydrogen quantity is less than 18 mol% in air, the detonation regime is not observed. Nevertheless, in this case, the flame speed can increase up to 70 or 80 % of the burned gas speed of sound. This flame acceleration has considerable effect on the confinement and can destroy the building. Flame acceleration has been largely studied for a homogeneous hydrogen distribution in the available containment volume [3-5]. Only very few data are available on the behaviour of the H_2/air flame in a non-uniform mixture [6, 7, 8]. The present work aims at providing an experimental database on flame acceleration of lean H_2/ air based mixtures characterized by hydrogen gradient. Experiments were carried out in a vertical large scale facility in order to obtain data to evaluate the key parameters to complete the criterion for gradients. The concentrations studied are within the range of 6 to 18 % of hydrogen in air and the characterization of the gradients after gas sampling is done through gas-chromatography.

2 Experimental layout

Experiments were performed in 5 m height vertical facility made of two distinct parts. The upper part is a 654 l vessel, named the dome, and it is connected to a vertical tube of approximately 65 l, referred to as the acceleration tube, in which 9 equidistant rings designed for optimum flame acceleration can be positioned (Figure 1). The internal diameter of the acceleration tube is 0.154 m and it is 3.3 m length and the dome has an internal diameter of 0.738 m and is 1.7 m long. The Blockage Ratio of the rings is equal to 0.63 ($BR=1-(d/D)^2$, where d is the internal diameter of the rings and D the internal diameter of the tube). This facility is highly instrumented with 15 photomultipliers tubes which allow the flame detection and 9 pressure sensors to measure the maximum pressure load. The dome and the acceleration tube are equipped with 3 optical windows each to allow laser diagnostics implementation.

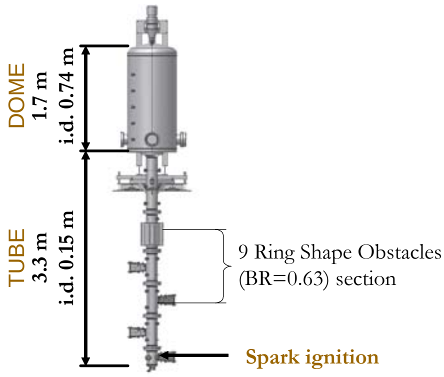

Figure 1: Sketch of ENACCEF.

The combustible mixtures were constituted of hydrogen distributed by Air Liquide (purity better than 99.95 %) and laboratory dry compressed air. Before each run, the whole facility was vacuumed down below

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1 Pa. Then, the mixture is introduced in ENACCEF via flow meter controllers (MKS1179A) at the desired composition up to a final pressure of 100 kPa. All the experiments were performed at ambient temperature.

The premixed gas at the desired concentration for the dome is introduced in the whole facility to reach approximately (depending on the desired final gradient) 89 kPa. The mixture is then only supplied from the bottom-end of the tube and the gradient is created by changing the concentration of the premixed mixture directly from the mass-flow meters. Moreover, the flow rate is sharply decreased (three times lower) to minimize mixing by convection inside the tube. A horizontal spiral with forty 0.8 mm holes oriented towards the bottom of the vessel is mounted to reduce the gas speed during filling. Both positive and negative gradients are created and a spark between two electrodes located at 0,138 m from the bottom of the vessel then ignites the mixture.

Gas sampling is made at six positions along the tube to characterize the gradient obtained prior to ignition. Gas chromatography equipped with a TCD detector is used to determine the concentration of hydrogen in the mixture and to control the ratio between N_2 and O_2 concentrations along the tube. An experimental procedure has been developed in order to be able to maintain a maximum error on hydrogen concentration less than 3% on the measurement value.

3 Results and discussion

After ignition, the spatial flame velocity is derived from the flame luminosity recorded by photomultipliers. For each configuration, several shots were carried out in order to obtain an average flame speed profile in the vessel and to assess the repeatability of the flame behaviour.The flame propagation has been studied in uniform mixtures as well as in presence of gradients of hydrogen in air. The effect of the hydrogen composition and distribution in the acceleration tube on the maximum flame speed has been experimentally investigated for both a smooth tube and a blockage ratio of 0.63. The measured gradient and the flame speed are plotted versus the distance from the electrodes. For all the experiments, the dome was filled with a uniform mixture. The gradient is created only in the acceleration tube.

Flame propagation in negative gradients

For the studied negative gradients, the flame accelerates in the obstacles area and can have different behaviours afterwards.

When hydrogen is distributed in the acceleration tube from 11.5 mol%, at the bottom, to 6 mol% at the top, the flame speed increases, within the obstacles field, before the flame is quenched (figure 2). The maximum speed registered for these experiments is around 265 $m.s^{-1}$ as it is shown in figure 2 (run 736, 737, 738). If the hydrogen concentration, at the ignition location, is increased from 11.5 mol % to around 16 mol% (run 740, 741), the flame propagates farther in the tube, the maximum flame speed was found to be around 700 $m.s^{-1}$, and the flame is quenched just before entering the dome as it is shown in figure 2. As it is shown in figure 3, the presence of obstacles enhances drastically the flame speed, since when the tube is smooth, the maximum flame velocity does not exceed 50 $m.s^{-1}$ while it reaches a maximum of 650 $m.s^{-1}$ as the blockage ratio is increased to 0.63.

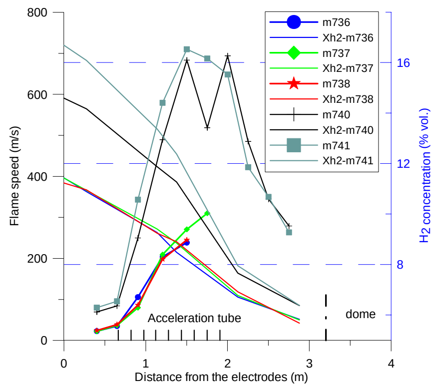

Figure 2: Flame speed and hydrogen concentration versus distance from ignition point in the case of obstacles with BR =0.63

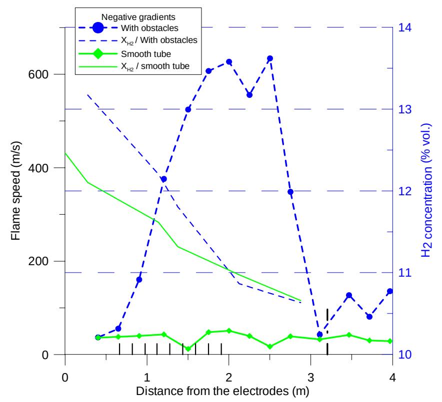

Figure 3: Flame speed versus the distance from the ignition point when the H_2 concentration varies from 13.17 mol% to 10.6 mol%, at a BR of 0.63 and from 12.5 mol% down to 10.6 mol% in the smooth case.

When the hydrogen molar fraction is kept around 6 % in the dome, and when the tube is obstructed, the flame is eventually quenched. This behaviour can be inferred to the effect of the turbulence created by the obstacles on the flame: the flame thickens and the structure of the flame front is a mixture between burnt and unburnt gases so the temperature of the reactive zone decreases and is not sufficient to allow propagation anymore. The activation energy for 7 mol. % H_2 (corresponding to an equivalence ratio of 0.18) is around 198 $kJ.mol^{-1}$ and 174 $kJ.\ mol^{-1}$ for 13 mol. % H_2.

Flame propagation in positive gradients

When the hydrogen content varies from 5.8 mol% at the ignition location towards 12 % at the top of the acceleration tube, the flame propagates to the entire volume (Figure 4). Maximum speeds reached are much lower than those observed with negative initial gradients (77 $m.s^{-1}$). Flame speed at first stage of propagation is difficult to measure since in such lean mixtures light emission is very low and the signal/noise ratio is high. Pressure signals and flame speed measurement in the dome prove that flame propagates to the top of the vessel. When the hydrogen percent is increased up to 10.5 at the ignition location (figure 5), the maximum flame speed increases up to 440 $m.s^{-1}$ in presence of obstacles and does not exceed 30 $m.s^{-1}$ in a smooth tube.

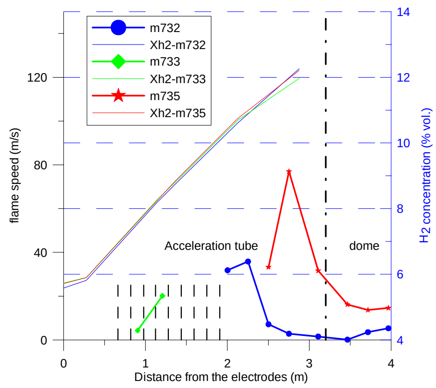

Figure 4: Flame speed and hydrogen concentration along the tube with obstacles (BR = 0.63)

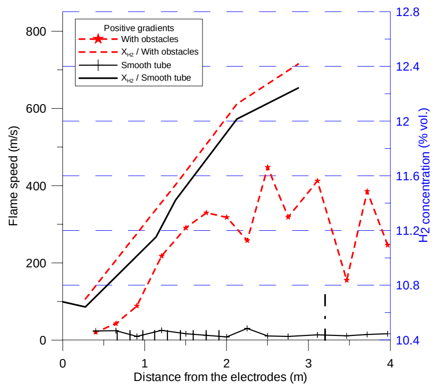

Figure 5: Flame speed and hydrogen concentration along the tube with and without obstacles.

Comparison with uniform distribution and acceleration criterion

For uniform mixtures a criterion has been built relying on the calculation of σ (the expansion factor), β (Zeldovich number) and Le (Lewis number) with the Chemkin package [9]. Three main cases were distinguished:

- Fast flames with a V_{max} / c_{sp} (ratio between maximum flame speed and the calculated sound speed in burnt gases) is around 0.5
- Slow flames when $V_{max} / c_{sp} \approx 0.3$
- Quenched flames

A previous study [8] has shown that a border separates the accelerated flames from the slow ones and is linked to β(Le-1) through the following equation: $\sigma^* = 0.075.\beta(Le-1) + 4.38$

For uniform mixtures with σ greater than σ* it has been shown that they have a very high propensity to accelerate. Is this criteria applicable for non-uniform mixtures? Which concentration should be taken into account in this case?

Maximum flame speed has been deduced from the experimental profile of the flame velocity and compared to the sound speed (in burnt gases) calculated for the average concentration in the tube (table 1).

$\frac{d[H_2]}{dx}(\%mol.m^{-1})$	**mol% H_2 Bottom → Top**	**$[H_2]_{average}$ (mol%)**	**V_{max} ($m.s^{-1}$)**	**C_{sp} ($m.s^{-1}$)**	**V_{max} / C_{sp}**
0	n.a.	10.5	260	671	0.387
0	n.a.	11.5	380	693	0.548
0	n.a.	13.0	535	725	0.734
-1.02	13.17→10.63	11,74	634	698	0.908
+0.676	10.70→12,42	11.63	448	742	0.604

Table 1: Maximum measured flame speed, sound speed in the burnt gas and their ratio for various compositions along the tube.

As its is shown in figure 6, the velocity profile depends strongly not only on the hydrogen content in the mixture (in case of uniform mixtures), but also when a concentration gradient exists on the sign of this gradient.

Indeed, when the gradient is positive, the velocity profile is comparable to the one observed in the uniform case containing the average value of 11.5 %. However, when the gradient is negative, the maximum velocity observed in the tube is even higher than the one obtained when the tube is filled with a uniform mixture of 13 % of hydrogen in air. In fact the flame behaviour seems to be driven by the H_2 content around the ignition location and the obstacles field. The parameters, σ and σ*, have been estimated for each concentration along the tube and plotted on figure 7. From this plot, one can see that in the case of positive gradients, the flame propagates in a mixture where the σ is lower than σ* in the acceleration tube and leads to a moderate acceleration of the flame. While in the case of negative gradients, the flame propagates, in the first half of the acceleration tube, in a mixture where σ is higher than σ*. Depending on the sign of the gradient, the "sigma" criterion cannot always be applied to the average concentration of hydrogen in the mxiture

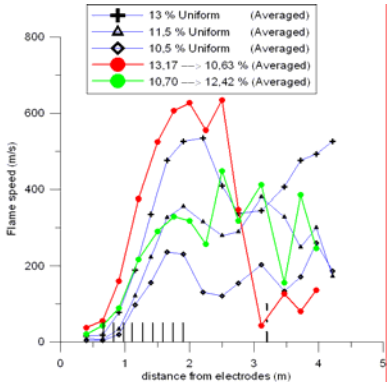

Figure 6: Comparison of flame speed between positive and negative H_2 gradients and uniform mixtures with obstacles (BR = 0.63).

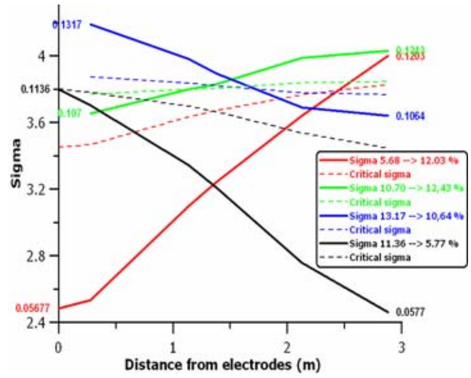

Figure 7: Expansion factor (σ) and critical expansion factor (σ *) versus distance from ignition point for various gradient.

4 Conclusions

This experimental work has been performed in order to assess the applicability of the "sigma" criterion to the assessment of the flame acceleration potential in case of non-uniform mixtures. During this work, it was shown that the flame propagation regime depends not only on the hydrogen distribution in the acceleration tube, but also on the sign of the gradient. It seems that the hydrogen concentration around the ignition and in the obstacles field is critical to the analysis of the flame acceleration. When the gradient is positive, the mixture seems to behave like the average one. When the gradient is negative, the flame propagates at a maximum speed that is even higher than the one observed in uniform mixtures containing the highest value of H_2 in air.

Acknowledgments

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