IR absorption measurements of the velocity of a premixed hydrogen/air flame propagating in a obstacle-laden tube

Roberta Scarpa, Etienne Studer, Benjamin Cariteau, Sergey Kudriakov,
CEA,
Saclay, France

Nabiha Chaumeix,
ICARE,
CNRS Orléans, France

1 Introduction and Motivations

In case of severe accident, in-core metal-water reactions due to fuel temperature increase lead to massive release of hydrogen into the containment of Light Water Reactors, as recently recalled by Fukushima accident. The safety features currently implemented for severe accident management cannot guarantee that the formation of large flammable clouds can be avoided. In case of hydrogen accumulation, a small amount of energy (such as an electrical spark) is sufficient to ignite the mixture. The explosion of such a cloud may eventually jeopardize the integrity of the containment and damage components important to safety or accident management. Moreover, the extent of this damage becomes more important as the propagation velocity of the reactive wave increases. The empirical flame acceleration criteria developed in the early 2000s by Dorofeev and colleagues [1] provide an effective tool for the analysis of the possible scenarios and it allows the selection of the most relevant situations for which flame acceleration may take place.

The validation of numerical tools capable to simulate the propagation of a premixed hydrogen/air flame in a large geometries is therefore a crucial issue for improving the safety of nuclear installations [2]. Industrial plants involving hydrogen in their chemical process or in their energy vector (Steam-methane reformer, fuel cell, etc.) deal with similar issues. This validation is generally based on few large-scale experiments and a large number of medium scale tube tests. This reduction of scale always enhances the number of sensors to measure the occurring phenomena. Hence, the most important effects can be identified and, as a consequence, the predictive capabilities of the models can be improved. For the tubes, these improvements are mainly the installation of visualization section [3]. Associated with Schlieren techniques and even with rapid field measurement techniques such as Laser Doppler Velocimetry (LDV), Particle Image Velocimetry (PIV) or Planar Laser Induced Fluorescence (PLIF), the carried out experiments have considerably increased the understanding of the acceleration processes of a premixed flame. Most of the experiments were generally

Correspondence to: roberta.scarpa@cea.fr

carried out with lean mixtures or at low initial pressure, or sometimes in an open tube. Nevertheless, these conditions do not cover all the accident-related situations and, parameters such as equivalent ratio, initial pressure and compression of the unburnt gases. Therefore, the use of tubes with small instrumentation ports remains of importance and raises the question of increasing the measurement capabilities of these devices. In order to measure the flame acceleration, the tube is usually equipped with photodiodes, photomultipliers tubes or ionization probes allowing the detection of the combustion wave at a point along the axis of the tube. The use of several devices along the path of the flame provides the spatial evolution of the 1D flame velocity. However, it is usually a very coarse representation with a maximum of a few dozen points. This technique proved to be sufficient to obtain the maximum velocity reached by the flame. But the increasing demands for validation processes require complete history of the transient related to flame acceleration.

Several efforts have therefore been made to conceive a system capable to measure velocity of the flame front in a continuous (non-discrete) way. Attempts have been made previously and there exists in the literature the works such as Kuznetsov et al [4] who used a photodiode to identify the time of the transition between deflagration and detonation. A measure by Oxygen light absorption was also recently implemented to record non uniformity of equivalent ratio in detonation tubes [5].

In this work we develop a method for extrapolating the velocity profile by measuring the variation of the extension in depth of the fresh (or burnt) gas along the tube axis. The method that we propose consists in performing IR-absorption measurements by doping the mixture with trace of alcanes such as methane. The advantages of using these compounds are 1) the small discrete region of absorption ($C-H$ bond in "'stretching mode"' between 2850 and 2960 cm^{-1}) and 2) the fact that they do only exist in the unburnt gas. Special care has been taken in correlating IR-absorption dependency on pressure and temperature variations. Eventually, comparisons between longitudinal and transversal flame velocity diagnosis techniques are drawn. From the literature [1] we know that, in an obstacle-laden tube, for a large spectrum of hydrogen concentrations, flame velocity increases rapidly up to a steady state value slightly lower than the Chapman-Jouguet deflagration velocity. Since our system is capable of capturing this fast increase with a high resolution (the recording frequency depends on HeNe laser-photodiode coupling) some progress can be done in understanding the dynamics of this phenomenon. Experimental results are then compared to the analytical ones proposed in the literature [14].

2 Measurement techniques and experimental apparatus

Since hydrogen/air mixture are unaffected by IR light, a small quantity of methane is added to the flammable mixture as tracer. The absorption in the infrared region of the O-H bond at 3.39μm is then used to monitor the flame velocity. This absorption technique has been employed extensively to measure hydrocarbon concentration in situ [6]. It foresses the use of a laser beam that is attenuated by the gaseous mixture. The transmittance τ of the gaseous medium is given by the Beer-Lambert law:

$$\tau = \frac{I}{I_0} = \exp\left(\frac{\sigma_i x_i P_0 l}{RT_0}\right) \tag{1}$$

with I the measured intensity, I_0 the light intensity throughout non-absorbing medium, x_i the molar fraction of the absorbing chemical species, P_0 the total pressure, T_0 the temperature, l the length of the seeded zone, σ_i the absorption cross-section and R the perfect gas constant.

The values for σ_i found in the literature [7–9, 11] have been verified in our laboratory at ambient temperature and pressure by using the device described in FIG. 1. Dependence of σ_i on the total pressure has been derived as well.

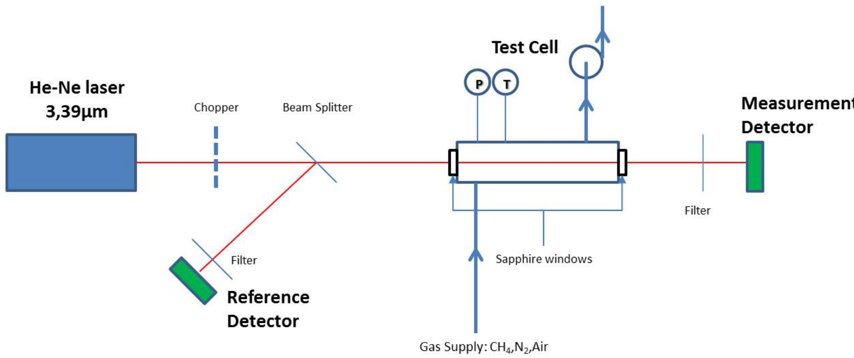

Figure 1: Small Cell used for Methane absorption determination

The apparatus, similar to the one used by Mevel et al [10] is shown in FIG. 1. The test section of total length of $l = 217$ mm is equipped with two sapphire windows, located at both ends of the tube that allow the laser beam to pass through the test section. The Helium-Neon Thorlabs H339P2 laser has a power of 2 mW and is chopped at 4000 Hz. Two Thorlabs PDA20H PbSe detectors (band width of 0.2Hz et 10kHz) were used to perform the intensity measurements. The results are given in Table 1 and Figure 2 and they are in accordance with the data found in the literature.

Table 1: Comparison of Methane absorption at $3,39\mu$m

σ_i (cm.atm)$^{-1}$ (at $P_0 = 1$ atm)	P_0 range	T_0 (K)	Reference
8	0,25 to 5 atm	298	[11]
9,2	10 to 1500 kPa	293	[7]
9,9	100 to 800 kPa	298	[8]
9,6	30 to 2000 kPa	300	[6]
8,2	500 to 2250 tor	302	[9]
8,2	1 to 11 bar	298	This work

Flame acceleration experiments of premixed hydrogen/air mixture were carried out in the SSEXHY (Structures Submitted to a EXplosion of HYdrogen) combustion tube of the CEA on the site of Saclay. This tube is made of three sections with a total length of $L = 3930$ m and an internal diameter of $D = 120$ mm is shown in FIG. 3. It is equipped with annular obstacles (blockage ratio BR $= 33$ % or 66%) located at one diameter distance along the path of the flame. The design pressure is 100 bar. Regarding the instrumentation, the tube is equipped with 42 openings (40 lateral and two axial) to accommodate sensors. The dynamic pressure is measured using Kistler 601A, 6001 and 7001 piezoelectric pressure transducers, the shock wave in the unburnt gas is detected using Chimiemetal piezoelectric sensors while the passage of the flame is recorded using Hamamatsu R11568 photomultiplier tubes. These devices are coupled with an optical system for collecting the light emitted by the OH radicals along the axis with a very narrow solid angle. For the IR absorption measurement, two sapphire windows 5 mm thick have been installed at both ends of the acceleration tube (FIG. 3) using the two axial openings provided with sapphire windows of 5 mm thickness.

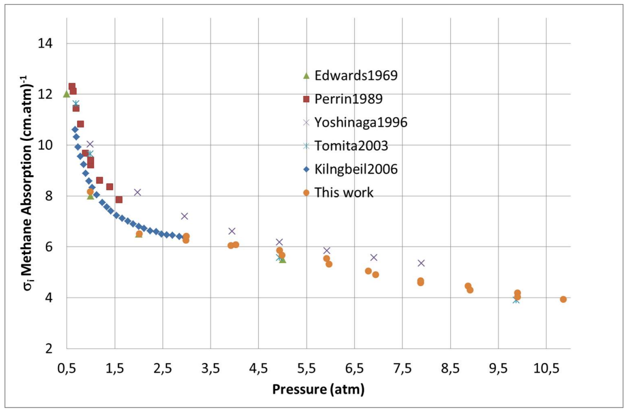

Figure 2: Effect of total pressure on Methane absorption cross-section at $3,39\mu$m

The initial length of the absorption zone section is $l = L = 3930$ mm. Since methane is consumed during the combustion process, length of the absorption zone evolves as:

$$l\left(t\right) = \frac{RT_0}{\sigma_i x_i P_0} \ln \left[\frac{I\left(t\right)}{I_0}\right] \tag{2}$$

assuming constant pressure and temperature in the unburnt gases.

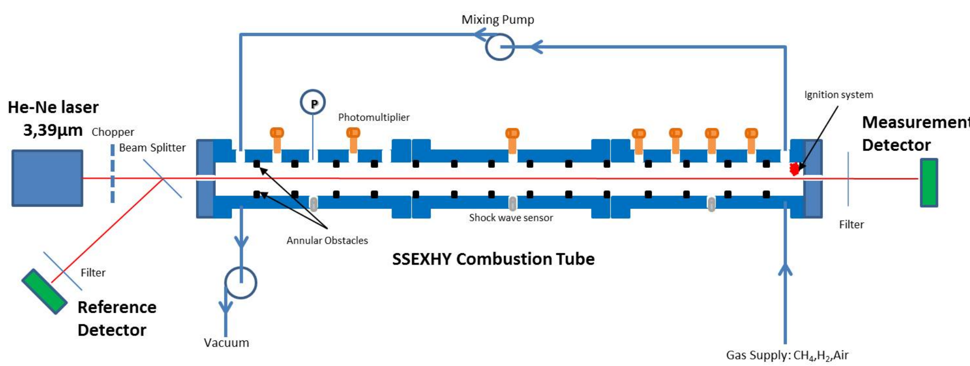

Figure 3: Schematic description of the SSEXHY facility

The gaseous mixture inside the tube is prepared by the method of partial pressures, starting by setting the tube under vacuum ($P < 2$ mbar), then injecting the methane (99.9995 % pure), the hydrogen (99.9991 %) and, finally, the air (20.9 % oxygen, 79.1 % nitrogen). A circulating pump is used for almost 30 minutes to homogenize the mixture inside the tube before ignition. At the end of the homogenization phase a gas sample is analyzed using a 490 micro-gas chromatograph (Agilent Technologies).

3 Experimental Results

Experimental results on flame acceleration using a 15 vol% of hydrogen in air mixture with an initial pressure of 1 bar at room temperature are given in FIG. 4. The main parameters of the combustion process are summarized in TAB. 2. The flame acceleration profiles are consistent with those found in the literature [12]; a chocked flame regime is reached at about 10 to 15 diameters depending on the blockage ratio (BR).

Table 2: Combustion parameters for hydrogen/air mixtures with 15 vol% of hydrogen at room temperature and pressure

Parameter	BR = 33%	BR = 66%
S_L (m/s)	0.78	0.78
σ	4.57	4.57
c_{su} (m/s)	375	375
c_{sb} (m/s)	760	760
V_{CJDF} (m/s)	722	722
V_{CJ} (m/s)	1524	1524
Re	150	110
a	2	4
b	1.5	1.5

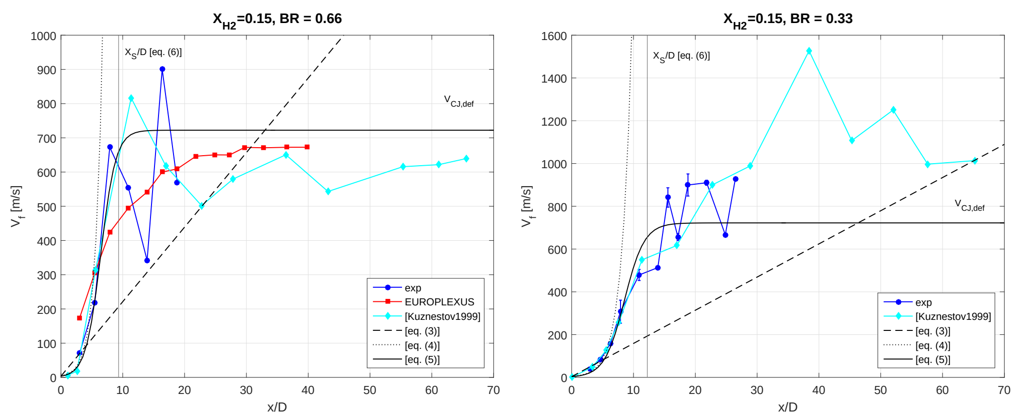

Figure 4: Left: Velocity profile for BR = 66% ; right: Velocity profile for BR = 33%

4 Discussions

The acceleration of a premixed flame in a tube with repeated obstacles has been theoretically described by Bychkov et al. [13, 14] under the hypothesis of incompressible flow in the unburnt and burnt mixture. The velocity of the flame tip is given in the form:

$$V_f = \frac{4\,(\sigma-1)\,S_L}{\sqrt{1-\mathrm{BR}}}\left[1+\frac{1}{2\,(\sigma-1)}\right]\frac{x}{D}+\sigma S_L \tag{3}$$

with σ the expansion ratio, S_L the laminar flame velocity and x the distance from the ignition point. This model only covers the initial phase of acceleration because the flame is assumed to be laminar, mono-dimensional in the tube and between the obstacles. However, this expression gives a first reference to compare our experimental results as can be seen in FIG. 4. Previously, Akkerman et al [15] proposed an analytical theory of flame acceleration in cylindrical smooth tubes in which the front accelerates exponentially according to the following expression:

$$V_f = (\sigma-1)S_L \exp\left(2\vartheta\frac{x}{D}\right)\frac{I_0(\eta)-1}{I_0(\eta)-2\eta^{-1}I_1(\eta)}+S_L \tag{4}$$

where $\vartheta = \frac{Re}{4}\left(\left(1+\frac{8(\sigma-1)}{Re}\right)^{\frac{1}{2}}-1\right)^2$, $\eta = \sqrt{\vartheta Re}$ and I_i the modified Bessel function of order i. Adjustment of Re number perfectly matches the first phase of the acceleration profiles (FIG. 4). Recently, Valiev et al. [13] proposed to extend their first model empirically by adding a term in order to reach asymptotically a chocked velocity:

$$V_f = \sigma S_L \frac{\exp\left(2\vartheta \frac{x}{D}\right)}{1+\alpha \mathrm{Ma}\sigma \exp\left(2\vartheta \frac{x}{D}\right)} \tag{5}$$

where $\mathrm{Ma} = \frac{S_L}{c_{su}}$ and $\alpha = \frac{S_L}{Mac_{sb}}$. This model is capable to qualitatively reproduce the measured acceleration profiles. Run-up distances X_S proposed by Dorofeev et al [16] have been added to the figures for comparisons.

$$\frac{X_S}{D} = \frac{a}{2}\frac{c_{sb}}{10S_L(\sigma-1)}\frac{1-\mathrm{BR}}{1+b\mathrm{BR}} \tag{6}$$

Finally, a numerical model of flame acceleration dedicated to large geometries has been developed [2, 17]. This simple model is based on the resolution of the Euler equations coupled to a diffuse interface propagation equation and the flame acceleration process is supplied by an algebraic equation of turbulent flame velocity. Results agree quite well with a number of flame acceleration experiments except for the initial flame development. The application of this model to the present experiments is also given in FIG. 4 (red line).

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