STUDY OF NITROGEN DILUTION, PRESSURE AND TEMPERATURE EFFECTS ON SPHERICAL FLAMES PROPAGATION OF $H_2/O_2/N_2$ MIXTURES

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1 Introduction

Since a few decades, there is an energy problem due to the depletion of the natural resources of coal, natural gas and oil. So, there is a need to find new sources of abundant and affordable energy. One of the foreseen solution reside in the fusion energy which led the international community to design the international scientific program ITER, for International Thermonuclear Experimental Reactor, to help in resolving this problem by producing large amounts of energy using nuclear fusion.

However, ITER operation may rise safety problems. Indeed, during the operation, plasma contained in the Vacuum Vessel (VV) erodes the surfaces of the walls composed by tungsten, beryllium and graphite. This plasma-wall interaction can produce several hundred of kilograms of metallic dust and graphite particles

In case of water or air ingress into the VV, when reaching high temperatures, steam may react with metallic dust and materials (beryllium, tungsten and carbon) on hot surfaces and produces hydrogen and carbon monoxide. After steam condensation, air ingress from the ex-vessel to the in-vessel break forms a flammable mixture, which can lead to high pressure loads in case of an explosion. The aim of the present work is to acquire data concerning the combustion of dust-hydrogen-air in a closed vessel, the first step being the study of gaseous $H_2/O_2/N_2$ mixtures.

These mixtures were already studied in the past [1-7]. Shebeko et al. [1] have worked on burning velocities and flammability limits of $H_2/O_2/N_2$ (or He or H_2O) mixtures at elevated temperatures (up to 523 K) and pressures (up to 4 MPa). Kishore et al. [2] studied the laminar burning velocity of H_2/O_2 mixtures diluted with $N_2/CO_2/Ar$ for a range (Φ=0.8-2.0) of equivalence ratios at atmospheric temperature (300 K) and pressure. Tang et al. [3] investigated the explosions characteristics (explosion pressure, combustion duration, maximum rate of pressure rise) of hydrogen-air mixtures diluted with nitrogen over a wide range of equivalence ratios (Φ=0.6-1.4) at elevated pressures (0.5 MPa) and temperatures (440 K). Tse et al. [4] have studied the morphology and the laminar burning velocity of spherical flames in H_2/O_2/inert gas mixtures at elevated pressures (up to 6 MPa). Qiao et al. [5,6] have worked on the laminar burning velocity and the Markstein number of H_2/air/diluent mixtures at different pressures (50 to 101 kPa) for several equivalence ratios (Φ=1 and 1.8). Aung et al. [7] have studied the effects of pressure and nitrogen dilution on the laminar burning velocity of $H_2/O_2/N_2$ mixtures for a wide range of equivalence rations (Φ=0.45-4.0), pressure (35-400 kPa) and volumetric

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oxygen concentrations in the nonfuel gas ($O_2/(O_2+N_2)$ = 0.125 - 0.210). In conclusion, if many data can be found in the literature concerning the laminar flame speed, one can find only the study of Tang concerning the maximum pressure rise rate and maximum explosion pressure

In this paper, $H_2/O_2/N_2$ mixtures combustion is investigated in a spherical bomb facility. The maximum combustion pressure (P_{MAX}) is measured and the maximum rate of pressure rise $(dP/dt)_{MAX}$ derived. Moreover, laminar burning velocity ($S_L°$) can also be extracted from the data of the experiments. Equilibrium calculations were made to obtain theoretical maximum pressure (P_{AICC} for Adiabatic Isochoric Complete Combustion Pressure) in order to compare with P_{MAX} and deduce the heat losses during the combustion. At the end, 0-D modeling calculations using COSILAB had been performed in order to compare $S_L°$ derived from the calculation to the experimental data. This comparison has been performed using two different detailed kinetic models.

2 Experimental setup and procedure

2.1 The Spherical bomb

The bomb is a stainless steel sphere (i. d. 476 mm) equipped with two opposites window (97 mm diameter, 30 mm thick); it has black polished surface. Two metallic electrodes located along a diameter of the sphere, are linked to a high voltage source. Ignition was produced at the centre of the sphere. The voltage and intensity discharge were measured with a high voltage probe and a current probe (fig. 1). The spherical bomb is equipped with a Kistler pressure sensor in order to measure the combustion pressure.

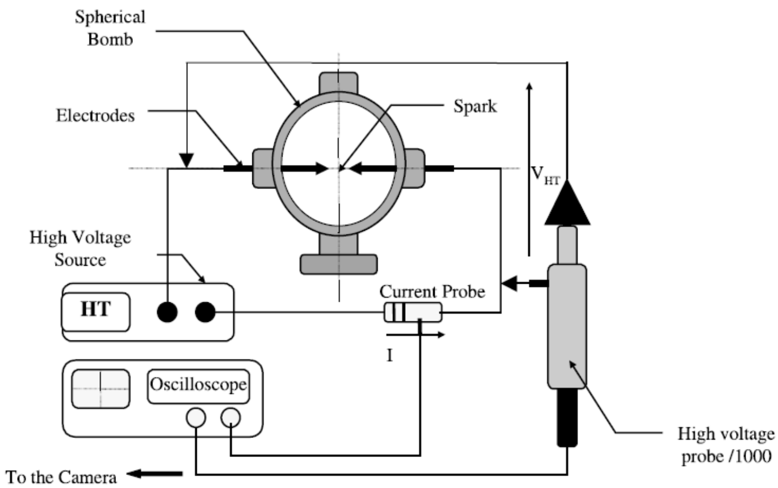


Figure 1. Scheme of the experiment

2.2 Flame visualization

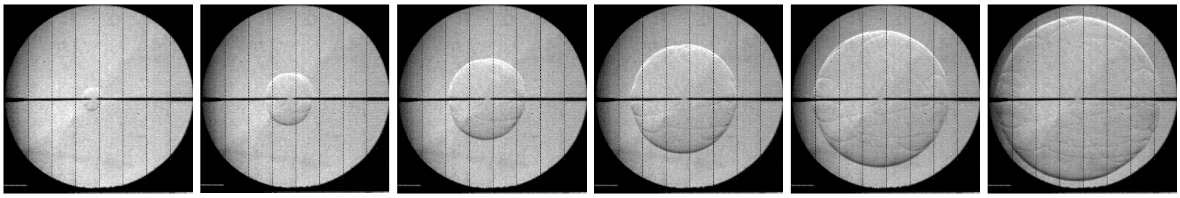
The visualization of the flame was obtained via a shadowgraph system. It consists of two concave spherical mirrors (focal length 1 m), the source light was a white continuous lamp and it was made as a point source via one bi-convex lens (focal length 20 mm). A numerical high speed camera with an acquisition frequency ranging between 2000 and 120 000 images per second was used to register the Schlieren images of the growing flames. These images allow the measurement of the radius of the flame as a function of time (fig. 2) using a homemade program [8].

2.3 Laminar burning velocity calculations

The spherical configuration, to study flames, has the advantage that the flame is well characterized through the experimental assessment of the stretch rate that is applied to the flame. The flame propagation is visualized via the recording of the flame front evolution as a function of time. When the observation is limited to the initial part of the flame expansion, where the pressure does not vary yet a simple relationship links the spatial flame velocity, V_S°, to the fundamental one, S_L° [9] (fig. 3):

$$S_L^\circ = V_S^\circ \cdot \frac{\rho_b}{\rho_u} = \frac{V_s^\circ}{\sigma}$$

where ρ_b is the burned gas density and ρ_u, the unburned gas one. The expansion factor, σ, is evaluated using the adiabatic flame calculations EQUIL of the Chemkin-II code package [10].



Frame 10 Frame 30 Frame 50 Frame 70 Frame 90 Frame 110

Figure 2. Schlieren images of the flame evolution in the spherical bomb. The mixture is constituted of 20% H_2 + 80 % air (21% O_2 + 79% N_2), initially at 50 kPa and 303 K. Framing rate: 15000 images/s

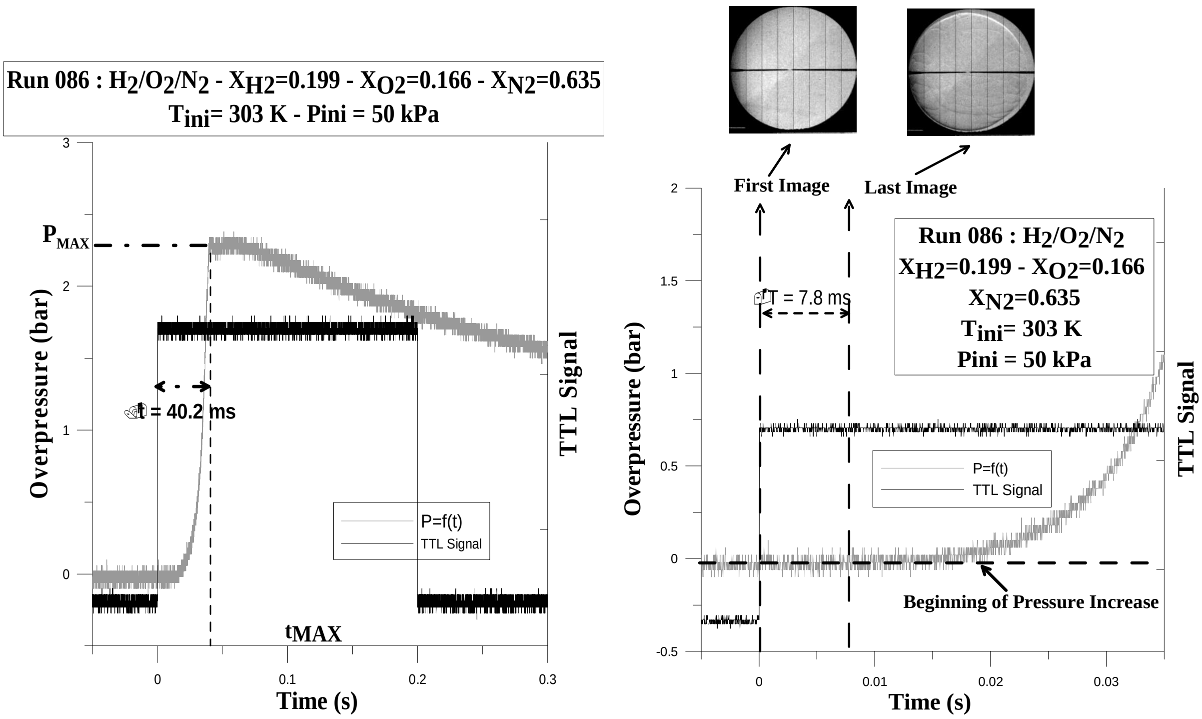


Figure 3. Pressure and TTL signals recorded during the test: (a): total time recorded. (b): zoom around the observation time. The mixtures was constituted of {20% H_2 + 80% {21% O_2 + 79% N_2}} initially at 303 K and 50 kPa

2.4 Experimental conditions

For this study, the hydrogen concentration has been varied over a wide range from 10 to 60 %vol, the initial temperature from 303 to 383 K and for the initial pressure from 50 to 300 kPa. The ratio of N_2/O_2 was varied from 0.66 up to 9.

In the following, hydrogen content reported are calculated by considering its volumetric fractions as follow: $\%H_2 = ([H_2] / [H_2]+[O_2]+[N_2])$. Oxygen content are oxygen volumetric fraction in N_2-diluted air and calculated as $\%O_2 = ([O_2] / [O_2]+[N_2])$.

The partial pressure method is utilized to fill the spherical bomb.

3 Results

3.1 Pressure results

The maximum pressure is an important parameter for the safety of the VV, we investigated the effect of mixture composition on maximum pressure achieved at the end of the combustion .Table 1 reports the maximum pressure observed experimentally (P_{MAX}) and the theoretical one (P_{AICC})for different mixtures containing 10% of H2 and for which O_2/N_2 was varied.

x% O_2	y% N_2	P_{MAX} (kPa)	P_{AICC} (kPa)	100 x (|P_{AICC} - P_{MAX}|)/ P_{AICC}
10%	90%	182.6	213.7±0.7	14.6%
21%	79%	188.5	211.4±0.4	10.8%
30%	70%	193.0	210.7±0.7	8.4%
40%	60%	192.9	209.7±0.1	8.0%
50%	50%	192.5	209.7±0.9	8.2%
60%	40%	189.3	208.1±1.4	9.0%

Table 1. Comparison of experimental and calculated pressure for different mixtures of 10% H_2 – 90 % $\{xO_2 + yN_2\}$ with various compositions of $\{O_2+N_2\}$ at ambient temperature and 50 kPa

Legend:

The relative error on the determination on P_{MAX} is estimated to be around ($\Delta P_{MAX}/P_{MAX}$) = 7%

As shown in table 1, for all reported conditions, the maximum combustion pressure is lower than the theoretical one by around 8 to 14.6%. Indeed, when the total volume is burnt, the maximum pressure recorded by the pressure transducer differs only by a few percent due to two phenomena: the heat loss by the flame to the walls and the Negative Flash Temperature (NFT) effect. When the flame reaches the sensor, it induces an increase in its temperature leading to a decrease of the voltage signal and thus artificially reduces the pressure. The ratio defined as (|P_{AICC} - P_{MAX}|)/ P_{AICC} ranges between 8.0 and 14.6% which gives an indication of the total amount of the heat losses and the NFT effect.

3.2 Maximum pressure rise rate

The maximum pressure rise rate $(dP/dt)_{MAX}$ is a safety key parameter for designing protection (safety) system. This parameter is deduced as a derivative pressure curve versus time at the inflection point.

As it is shown in figure 4, the maximum pressure rise rate is nearly constant and has a value of 19±1 bar.s^{-1} for a large range of N_2/O_2, namely between 0.66 and 3.76. Nevertheless, this value becomes lower than 12 bar.s^{-1} for large value of N_2/O_2. When N_2/O_2 is lower than 4, the maximum pressure rise rate depend weakly on this ration as both the maximum combustion pressure (cf. Table 1) as well as the total time of combustion are very close (479 ± 41 ms). While when N_2/O_2 is equal to 9, the combustible mixture is highly diluted by N_2 leading to a much longer combustion time (675 ± 46 ms).

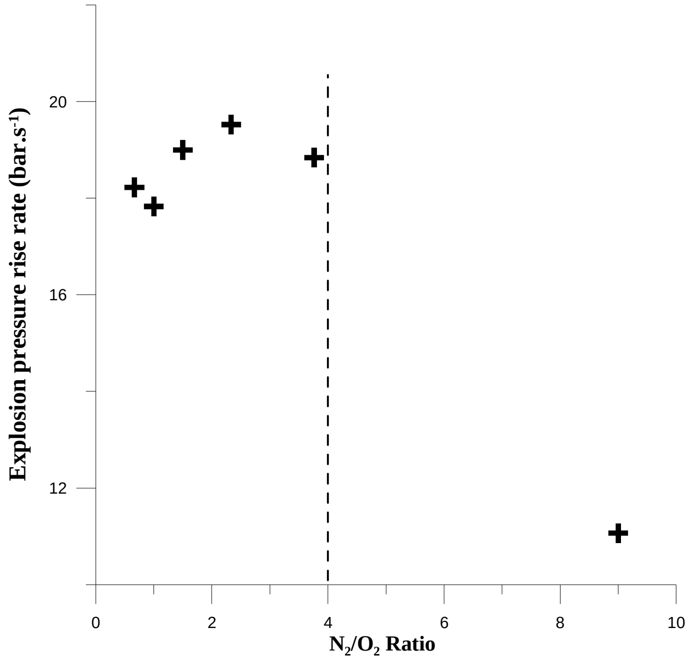


Figure 4. Explosion pressure rise rate of 10 % hydrogen - 90% {xO_2 + yN_2} mixtures versus N_2/O_2 ratio at ambient temperature and 100 kPa.

3.3 Laminar burning velocity and modeling

The laminar burning velocity of the different mixtures has been evaluated experimentally and compared with two detailed kinetic models in order to choose which one will be taken to better represent the experiment. The calculations are made with the software COSILAB and the premixed, freely propagating flame model. The models studied in this work from Wang et al. [11] and Mével et al. [12] which contains respectively 8 species for 28 reactions and 32 species for 203 reactions. The main difference between these two mechanisms is the presence or not of nitrogen species in the combustion process.

% O_2	% N_2	Vs° ($cm.s^{-1}$)	Sigma (σ)	S_L° (cm/s^{-1})	S_L° (cm/s^{-1}) Wang Model	S_L° (cm/s^{-1}) Mevel Model
10	90	394.45	4.2765	92.24	90.01	98.43
21	79	554.91	4.9080	113.06	114.86	118.84
30	70	631.58	4.8950	129.02	119.07	121.61
40	60	705.08	4.8854	144.32	120.63	122.38
50	50	716.20	4.8627	147.28	120.72	122.00
60	40	711.41	4.8359	147.11	120.37	121.32

Table 2. Comparison of experimental and calculated laminar burning velocity for different mixtures of 20% H_2 – 80 % {xO_2 + yN_2} with various compositions of {O_2+N_2} at 343K and 100 kPa

The first results show that Wang and Mével models give similar results for all the cases studied here. The two models are quite representative of the experiment for normal air composition but underestimated S_L° for the mixtures with 'air' enriched with oxygen. But, we have to remark that the mixtures studied here give cellular flames. So, the experimental spatial velocity may be overestimated.

4 Conclusion

Data has been obtained on laminar flame speed and maximum combustion pressure for $H_2/O_2/N_2$ mixtures using the spherical method. The maximum combustion pressure is independent of N_2/O_2 ratio in the case of complete combustion for very lean mixtures. The maximum pressure rise rate is constant for a N_2/O_2 ratio between 0.66 and 3.76 and decrease sharply for a N_2/O_2 ratio equals to 9. The laminar flame speed modeling is still under process.

Acknowledgment

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