

## IGNITION EXPERIMENTS OF HYDROGEN MIXTURES BY DIFFERENT METHODS AND DESCRIPTION OF THE DRDC TEST FACILITIES

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### ABSTRACT

The paper will present results of hydrogen/oxygen mixtures ignited by using electric sparks, electrostatic discharges, a heating element and a flame. Measurements of the lower flammability limit (LFL) was done for each ignition method. The hydrogen mixtures of different concentrations were ignited at the bottom of a combustion chamber, leading to an upward propagation of the resulting flame. At some level of concentration the combustion was partial due to the limited upward propagation. The complete combustion of the whole mixture was observed at concentration limits higher than the known LFL of 4% vol. for hydrogen in air. The paper will describe the test facilities and the resulting ignition probabilities for different ignition methods.

## INTRODUCTION

Hydrogen mixtures with pure oxygen or air may be ignited by different processes such as electric spark, electrostatic discharge, thermal ignition (heating element, open flame, incendiaries, autoignition temperature...) as well as by shock wave, mechanical impact and by a rapid adiabatic compression.

Ignition of hydrogen/air mixtures were done at various concentrations inside a plastic bag. It was done in order to verify if the hydrogen would completely combust at concentration ranging around the low flammability limit (LFL). Ignition of hydrogen mixtures was also done with other ignition processes inside of a stainless steel chamber to observe the differences in the efficiency of hydrogen combustion.

## 2.0 EXPERIMENTAL SETUP

### 2.1 Ignition experiments setup (lower flammability limit)

The setup designed to perform Lower Flammability Limit (LFL) measurements was based on instrumented hydrogen bag shown in Figure 1 and was composed of the following items described on Figure: a) temperature and thermal flux acquisition system, b) electric spark plug, c) hydrogen valve, d) hydrogen detector inside the bag e) visible and IR light cameras (not shown on Figure). LFL is the lowest concentration at which a fuel is expected to ignite and propagate the flame. A data acquisition system was developed and adapted with *labview* software to perform measurements of various parameters that can be reported on the same diagram. The acquisition system simultaneously acquires and reports the following parameters in a single Figure: a) hydrogen concentration, b) temperature and c) recording of the burning process by two cameras (IR and visible). The acquisition speed of the following parameters was 50 points/sec: temperature, thermal flux and hydrogen concentration. The data recorded by the system were then averaged for each second by the data treatment process.

It was important to find an appropriate plastic material that did not generate a good amount of IR light. Transparency property of the material to IR emission was also important. These properties of the material were essential to be able to record hydrogen ignition by the IR camera. After few trials with some different types of plastic materials, it was found that Polyvinyl Fluoride "*Tedlar Dupont*" plastic bag did not generated IR emission and was also transparent to IR light emission. The material was also able to sustain hydrogen air mixture for more than 5 minutes without any major leaks. The maximum volume that may be injected inside of the bag was 5 L.

Few types of IR cameras were tested to maximize the signal coming from the ignition of the hydrogen/air mixtures. It was found that the 3-5  $\mu\text{m}$  IR camera was the best system to observe the burning inside of the plastic bag. The speed of acquisition of both cameras (IR and visible) was 30 frames/sec. Recording of the temperature inside the bag was an important parameter that gives an idea about the ignition of the hydrogen/air mixture. It was performed to evaluate conditions leading to ignition of hydrogen mixtures and the low flammability limits.



Figure 1 – Instrumented Hydrogen bag: A) thermal flux sensor, B) spark plug, C) hydrogen valve, D) Hydrogen detector

## 2.2 Measurement of the energy of the electric spark

An electric spark generator was mounted in the laboratory. It was composed of a trigger button, a power supply and an accumulator. The distance between the electrodes of the spark plug was measured to be 0.50cm (0.20 in). It is understood that the distance between the electrodes could influence the energy of the electric spark produced. Ignition of the hydrogen mixture using the spark plug system could be synchronized with the data acquisition process. Appropriate safety procedures were established. Since a direct measurement of the initiation spark energy is rather difficult, we resorted to an indirect measurement of the electrical energy dispensed to the spark. The spark duration was measured by looking at the light pulse generated by the spark with a 20 mm<sup>2</sup> silicon PIN photodiode (model ET 4000 from Electro-Optics Technology) placed at about 20 cm from the spark and coupled to a TDS 3054b Tektronix oscilloscope. The measured FWHM pulse duration was 36 ns.

The spark initiator was driven by a 24 V dc supply as shown on Figure 4 and had a trigger button, which, when activated, results in a continuous stream of 36 sparks per second. Under this condition the mean current taken by the spark initiator is 20 mA so that the maximum energy available per spark amount to 13.3 mJ. The overall efficiency of the spark initiator circuit is unknown and likely to depend on the spark length and the surrounding gas composition but value of between 0.2 to 0.75 seems likely. In this case the initiation energy would fall between 2.5 and 10 mJ.

### 2.3 Measurements of the lower flammability limit (LFL)

Experiments on the lower flammability limits of hydrogen/air mixtures were performed inside the 5 L plastic bag using the ignition experimental setup described above. The bag was filled with 2 L of hydrogen/air mixtures to avoid overpressure during ignition experiments. The criteria that defined the ignition of the hydrogen/air mixture were the observation of the flame by the IR camera and the instantaneous change of the temperature after the flash. The ignition was also leading to an additional inflation of the bag observed by the visible camera as well as a signal observed by the thermal flux sensor.

Dispersion of the flame inside of the bag can be easily observed as shown in Figure 1a to 1d. The ignition was initiated at a central point of the bag. The drop of the hydrogen concentration was measured during the combustion process. In some cases it was found that the combustion was partial, leading to residual hydrogen concentration after the combustion of the mixture had stopped.

A hydrogen/air mixture of around 8% vol. was ignited by triggering the electric spark and the combustion process is shown in Figure 1b. Combustion resulted in a drop of the hydrogen concentration to 4.3 %vol. It demonstrates that the hydrogen did not burn completely inside the bag as shown on Figure 1b and that only the hydrogen at the upper part of the bag ignited. The bag was filled again with a hydrogen/air mixture up to 5.0 % and ignited again by triggering the electric spark. The mixture partly burned. In all cases it seems that the combustion stopped at hydrogen concentrations around 4.3% vol. inside the bag. Upward propagation of the flame was observed by the IR camera, while the hydrogen contained in the lower part of the bag did not ignite as shown more clearly in Figure 1c and 1d with hydrogen concentrations below 8% vol. Consequently, it is suspected that downward flame propagation would have also consume all the hydrogen contained inside the bag at concentrations higher than 9.5%.

Peak temperature produced by the flame inside the bag depended on the initial concentration of hydrogen. The peak temperature inside the bag was 34°C with an initial hydrogen concentration of 8.0% vol, while the peak temperature was only 24.3°C at an initial hydrogen concentration of 5.5% vol. Peak temperature for mixtures of 11.5% vol. was observed to be 53°C. It is obvious that peak temperature may vary significantly depending on the volume and the concentration of the ignited hydrogen mixture.

Numerous measurements were performed to evaluate the probability of ignition of different hydrogen concentrations ranging from 4.0 to 6.0% vol. Results include 125 experiments. The hydrogen was mixed with air by using a syringe. No cases of ignition were observed at a hydrogen concentration of 4.9 % vol. inside the bag. It represents the LFL value of a hydrogen/air mixture at 23°C and atmospheric pressure. The value observed with our experimental setup is slightly different from the known LFL of 4.1%, probably due to the fact that the flame was not easily observed at this low concentration and the hydrogen concentration drop that might have occurred during combustion was below the sensitivity of the sensor.

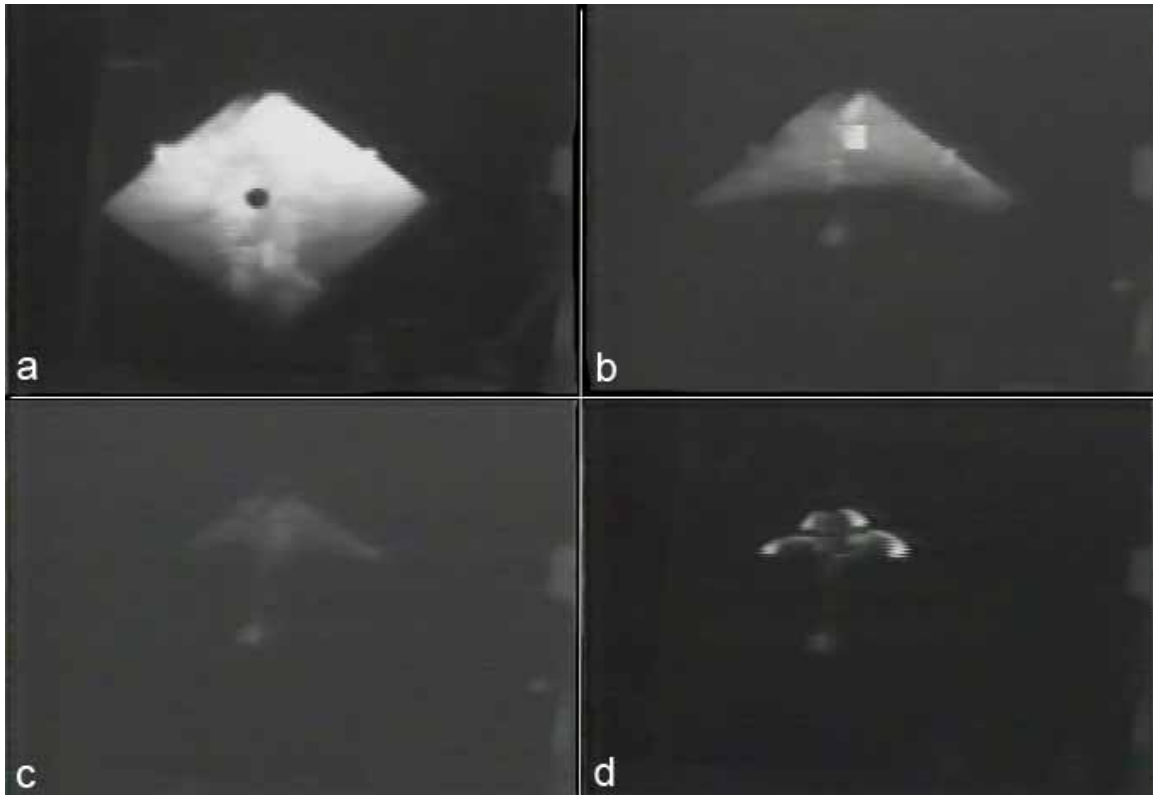


Figure 1 – Ignition of hydrogen air mixtures for different concentration ranging from 10% down to 5%

Concentration leading to a complete combustion of the hydrogen mixture was determined. The results taken from numerous ignition experiments are summarized in Figure 2. It shows that the amount of hydrogen necessary to burn all the mixture was around 9.6 %  $H_2$ . The value is different from LFL and is coming from the fact that the hydrogen concentration leading to a downward propagation of the flame is different that of an upward flame propagation. This result is consistent with earlier reported in the literature results for downward flame propagation between and 9 and 10% vol. [1].

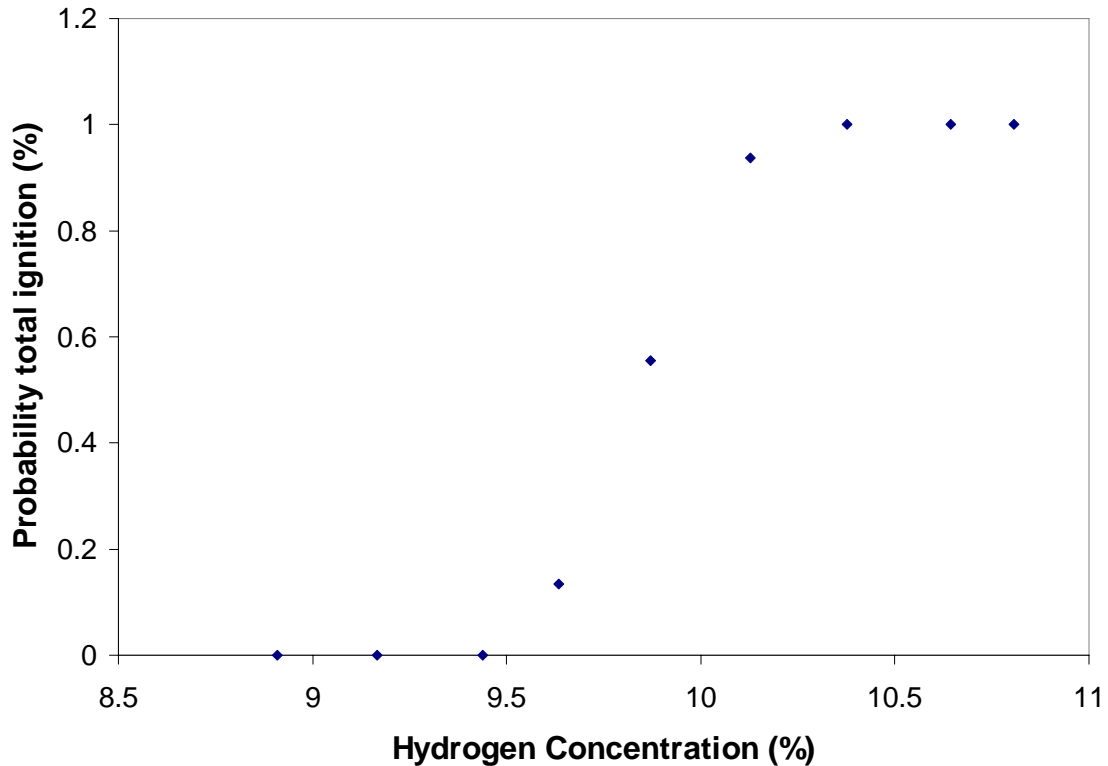


Figure 2 – Probability leading to a complete combustion of all the hydrogen contained in the bag vs the concentration of the hydrogen/air mixture

#### 2.4 Thermal flux measurements inside the bag

Measurement of the thermal flux ( $\text{W}/\text{cm}^2$ ) inside the bag was made possible during the ignition of hydrogen mixtures. The thermal flux sensor model HFS-4 from *Omega co* was assembled with appropriate measurement equipment. The sensor was based on measurement of temperature differences between two thermocouples attached into a thin polymer matrix. The sensor is designed to measure heat flux to or from a surface with minimum disturbance of the existing heat flow pattern. The sensor is of a temperature gradient (Schmidt) type in which a self generating thermopile is arranged around a thin thermal barrier to produce a voltage that is a function of the thermal energy passing through the sensor. Electrical signals coming from the thermal flux sensor were calibrated and measured to be between 0.006-120 millivolts, depending on the intensity of the heat flux. The range of thermal flux that could be measured by the sensor was between 0.03 milliwatt/  $\text{cm}^2$  to 0.63  $\text{W}/\text{cm}^2$ . The sensor was attached to the inside layer of the bag to make it work. Thermal flux measurements were to give an idea of the heat and the intensity of the flame produced.

This can be used to evaluate potential harm to human skin exposed to the heat produced by the flame. Risk of injuries may be evaluated by using known values. For example, it is recognized that human may feel pain in 15-30 sec with a thermal flux of  $0.47 \text{ W/cm}^2$ , while skin will burn to a 2th degree will burn in 30 sec. A thermal flux of  $0.95 \text{ W/cm}^2$  would give an instantaneous skin reaction, while skin will blister after 5 seconds at  $1.6 \text{ W/cm}^2$ . Polywood and hardboard will ignite with a thermal flux of  $1.0\text{-}1.8 \text{ W/cm}^2$  after an exposure time from 60-300 sec. A safe limit for extended exposure of humans is recognized to be  $0.16 \text{ W/cm}^2$ .

Thermal flux measurements from ignited hydrogen/air mixtures were performed inside of the plastic bag. Values observed for the ignition of hydrogen/air mixtures was of  $0.42 \text{ W/cm}^2$  for a hydrogen concentration of 10% and  $0.08 \text{ W/cm}^2$  for a hydrogen/air mixture of 8.5%. The fast burning time and the weak thermal flux conditions inside of the bag were not high enough to cause injuries to the skin.

## **2.5 Electro Static Discharge (ESD)**

Ignition experiments were performed by using an electro static discharge system that was mounted in the lab. The configuration of the electrodes inside the bag is described in Figure 3. The ignition of the hydrogen mixtures were observed through a window by using the IR camera.

The discharge was produced by a capacitor charged to a high voltage. The energy of the capacitor  $E$  (Joule) is related to the capacitance  $C$  (Faraday) and the voltage  $V$  (Volts) by the following equation:  $E = 1/2 CV^2$ . The hydrogen mixtures were ignited at 25kV using a capacitor of 540pF. The voltage drops was observed during the discharge. The duration of the discharge was about 0.4 sec.

Numerous ignition trials were performed at different concentrations. It was deduced based on probability of ignition that the LFL of hydrogen/air mixture from electrostatic discharge was 3.9%.



Figure 3 – Setup used to ignite hydrogen inside a bag using a calibrated electrostatic discharge device.



## 2.6 Ignition using a red hot element

Hydrogen mixtures were ignited inside of a stainless steel chamber by using a red hot element. The temperature of the red hot element was calibrated by applying different voltages and by using a thermocouple touching the element. Consequently it was possible to determine the concentration as well as the lowest temperature of the red hot element leading to the ignition of all the hydrogen contained. Hydrogen mixture could be ignited with the hot element at  $375^{\circ}\text{C} \pm 25$ . The mixture could not be ignited below that temperature. This temperature appears to be substantially lower earlier reported self-ignition temperature of hydrogen in air: between 520 and 585 C depending on the literature source [2]. The difference could be attributed to the experimental set up that took the temperature measurements from the red hot element but not at the point of contact with the gas mixture.

The heating element was at the upper part of the chamber. So it could ignite the hydrogen mixture by the downward propagation process. The ignition was observed by measuring the change of the hydrogen concentration and the increased of the temperature produced by the burning process. These sensors were at the bottom of the combustion chamber. A partial combustion inside the chamber was not leading to a major increase of the temperature as well as a significant consumption of the hydrogen observed by the sensor. Numerous ignition experiments were performed at different concentrations. The resulting flammability limit was considered to be the concentration leading to a total combustion of the hydrogen inside of the chamber. It was observed that the hydrogen completely burned by a downward propagation mechanism at 9.6 % vol.

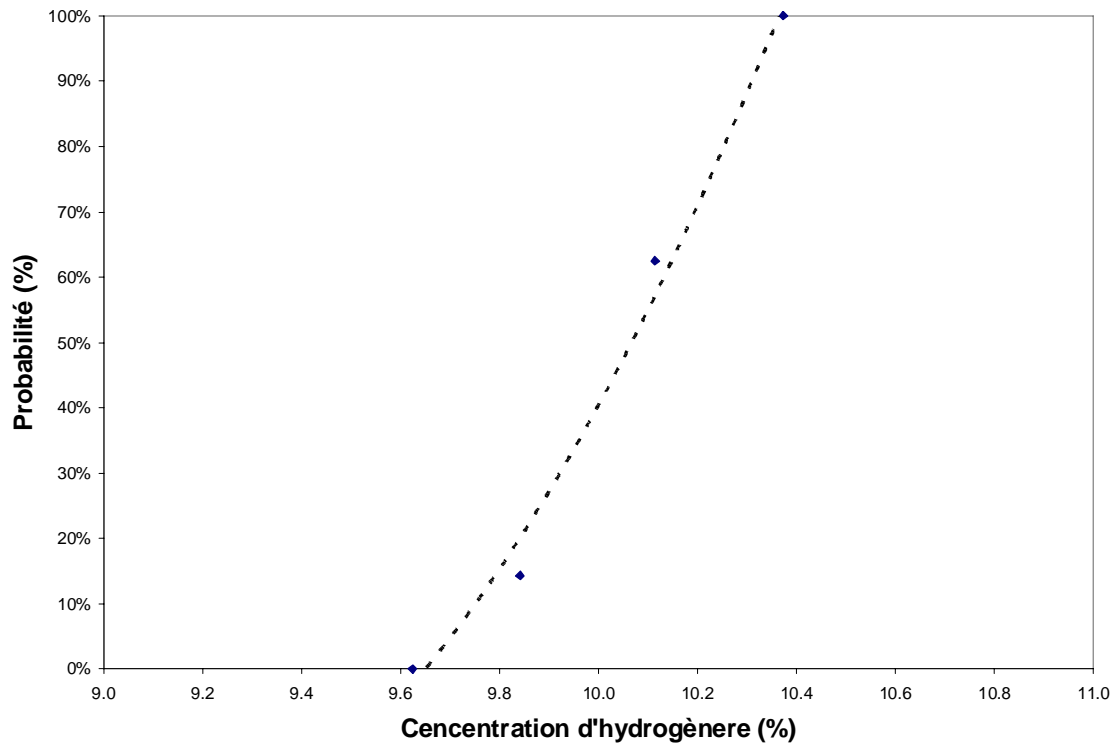


Figure 4 – Probability of complete combustion of hydrogen/air mixtures using a red hot element

## 2.7 Ignition by a flame

The stainless steel chamber was used to measure the ignition of hydrogen/air mixtures. A device that remotely controlled the movement of an ignited candle was designed. The ignition of the hydrogen mixtures was observed by the IR camera as shown in Figure 4. The picture on the left of Figure 4 is showing the open flame just before the ignition of the hydrogen, while the picture on the right (Figure 4) is showing the partial ignition of the hydrogen mixture while a total burning of the hydrogen contained inside the chamber is observed in Figure 5 (right picture). The concentration that has zero probability to ignite the mixture was determined by numerous experiments. The results are shown on Figure 6. Total ignition of the whole mixture was observed at concentration higher than 9.2%, as shown on Figure 5 and 7. In this case the burning of the hydrogen was complete and was clearly leading to a much more violent flame. The concentration of the complete combustion inside the chamber corresponds to the same value as what was observed for the ignition using the red hot element. It corresponds to a lower flammability limit burning process produced by a downward propagation mechanism.



Figure 4 – Partial combustion of hydrogen mixtures between 4.2% and 9.4%



Figure 5 – Complete combustion of hydrogen mixtures at concentration higher than 9.4 %

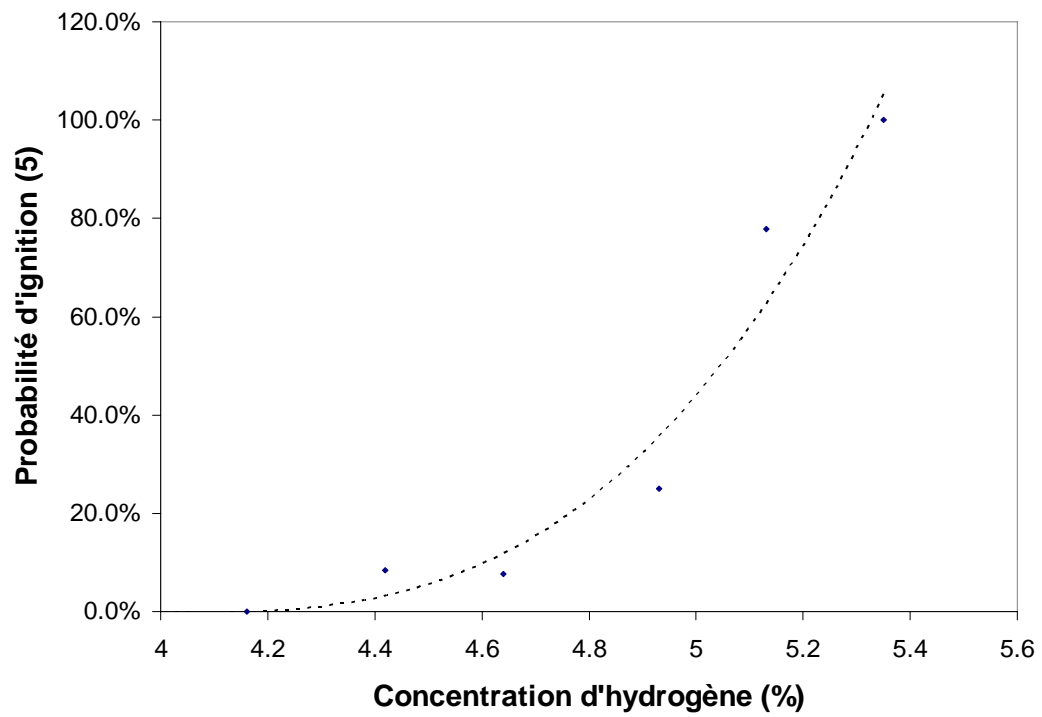


Figure 6 – LFL measurements for the ignition by a flame

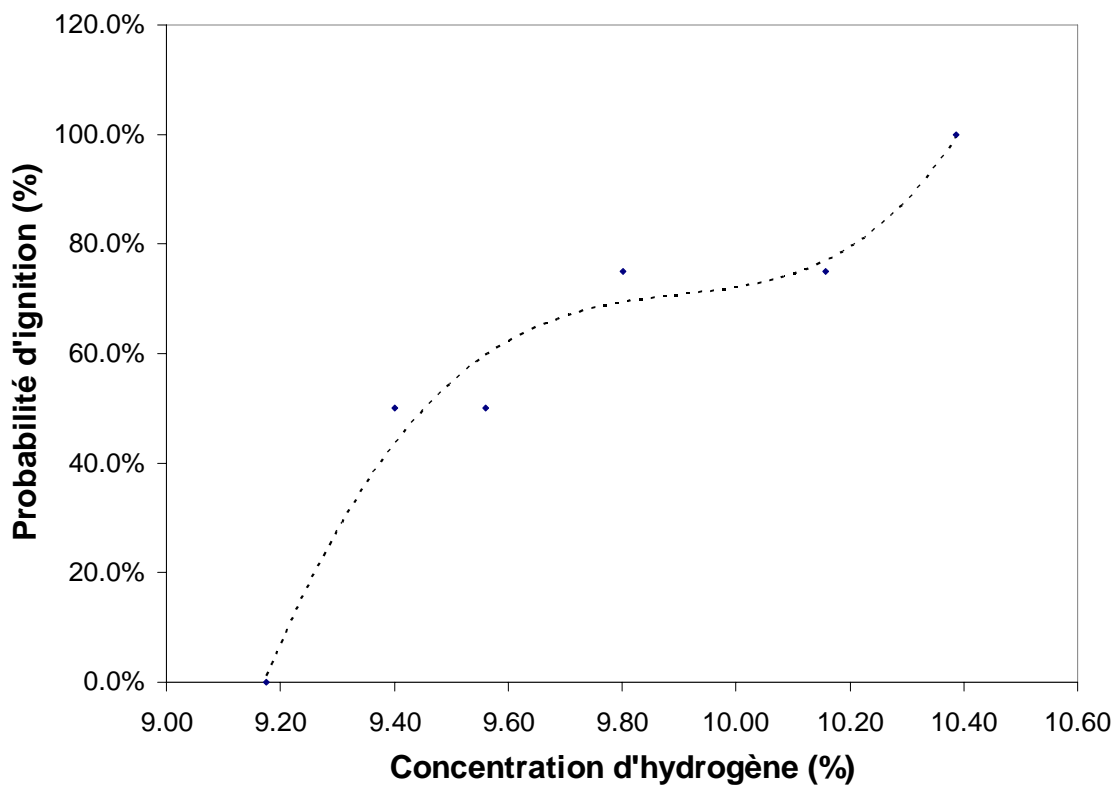


Figure 7 – Total probability of complete combustion of a hydrogen/air mixture

## 2.8 CONCLUSION

Ignition of hydrogen/air mixtures were made in different concentrations with four types of ignition process. Measurements of the low flammability limit were found to be very similar for every ignition mechanism. Experiments showed that the hydrogen/air mixture may not burn completely at the LFL value unless it is ignited at the very bottom. This was explained by the fact that LFL concentration may sustain only the upward flame propagation process that may not lead to a complete combustion of the hydrogen/air mixture.

## 2.9 ACKNOWLEDGEMENTS

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## REFERENCES

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