A new technique to produce well controlled electrical sparks. Application to MIE measurements

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Abstract

Few laboratories in the world have kept the capability of measuring minimum ignition energies (MIE). When a flammable gas needs to be allocated an ATEX group other measurements may be performed like determining the minimum experimental safety gap (MESG). However, the relationship between MESG and MIE is statistical and certainly lacks accuracy and when more accurate information is required as for instance when investigating the influence of the initial conditions on the ignitability, direct MIE measurements are required. To be able to meet such request, the author's team re-developed an expertise in this field be using up to date technologies. A brief review of the methods used in the past is first given highlighting the difficulties. Then a new technique is described with which the losses can be accurately estimated. The method was used for propane and hydrogen air.

Keywords: *minimum ignition energy, spark ignition*

1. Introduction

To implement safety in the process industries and to market flammable goods, accurate safety data are compulsory. As far as explosible mixtures are concerned, some key parameters are the minimum ignition energies (MIE), the minimum auto ignition temperatures (AIT) and the flammability limits (FL).

An intense effort was devoted by the industrialized countries during the XXth century, primarily to better define these parameters and to develop adequate testing methods, secondly to relate them to the practical ignition circumstances and thirdly to organize the market of the flammable goods (REACH directive) and of equipments to be used in dangerous areas (ATEX directives). But it seems that the technical and scientific effort related to the physics of ignition has dropped since 2 decades, the concerned laboratories having switched their core activity towards certification. Nevertheless, new flammable goods appear for which the well accepted experimental methodologies for traditional fuels may not be suited (for instance of too small size if the product is slightly flammable) and would require at least a critical appraisal.

The aim of this work is to revisit Minimum Ignition Energy measurements and (re)develop an expertise especially in view of investigating very sensitive mixtures. Below are recalled the main physical aspects and data about spark ignition. Then the technology developed is presented and results are shown and analysed.

2. Spark ignition

2.1. Some theoretical considerations

A theoretical "spark" could be defined as a punctual and instantaneous deposit of energy. Good monographs have been published along the last 50 years regarding the underlying physical mechanisms of flame development including ignition processes (Lewis and von Elbe, 1987; Glasmann, 1977; Williams, 1985; Hattwig and Steen, 2004). Extending the work of Brokaw and Gerstein (1957), the classical "thermal explosion theory" can be used to describe the phenomenology and this shows the links between the ignition and combustion parameters. Most analytical developments rely on a highly simplified chemistry and, often, the heat release is represented by a one-step chemical reaction obeying a global Arrhenius reaction rate:

F (fuel) + O (oxidant as air) -> P (products) + heat

With the volumetric heat release rate, Q_{comb} (W/m^3) in a simple form :

$$\dot{Q}_{comb} = \rho \cdot \Delta H_{comb} \cdot A \cdot e^{-\frac{E_{act}}{R \cdot T}} \qquad [1]$$

Where:

- ΔH_{comb} is the specific heat release rate per unit mass of the mixture (J/kg)
- ρ the specific mass of the mixture (kg/m^3)
- A the preexponential factor of the Arrhenius law (s^{-1})
- E_{act} the energy of activation of the reaction (J/mole)
- R the perfect gas constant (8.314 J/mole/K)
- T the temperature (K)

Note that ΔH_{comb} is a property of the mixture and is related the maximum temperature of the combustion (T_{ad}) as measured in an adiabatic system. If T_0 is the initial temperature, ρ_0 the initial specific mass and Cp the specific heat capacity then [1] reads:

$$\dot{Q}_{comb} = \rho_0 \cdot A \cdot Cp \cdot (T_{ad} - T_0) \cdot \frac{T_0}{T} \cdot e^{-\frac{E_{act}}{R \cdot T}} \qquad [2]$$

Because E_{act}/R is typically on the order of 10000 K for most fuel-air mixtures, the evolution of [2] with T is largely dominated by the exponential term as shown on figure 1. Note also that this equation is valid for T smaller or equal to T_{ad}. This expression does not include any limitation so that it may be inferred that combustion would occur at any temperature even without any ignition source. The speed of the combustion process would only depend on T. This might be true only if the process was perfectly adiabatic but not when heat losses are taken into account. Unless T is very large, gases exchange heat mainly by conduction and convection. Assuming a volume V (m^3) of the gas having a typical size D (in m, for instance $D = V^{1/3}$), then the volumetric heat lost by conduction or convection with the surrounding (Q_{comb} in W/m^3) through the external area $A_{exchange}$ (m^2) of V reads:

$$\dot{Q}_{losses} = h_{conv} \cdot \frac{A_{exchange}}{V} \cdot (T - T_0) \approx h_{conv} \cdot \frac{1}{D} \cdot (T - T_0) \qquad [3]$$

where h_{conv} is the exchange rate coefficient (unit of W/m^{2}K), usually approximately constant for a given geometrical configuration and flowing conditions. It appears that, for a given size D, [3] varies linearly with T as shown on figure 1. Equations [2] and [3] then describe the behavior of a pocket of reactant raised at a given temperature T (Fig. 1). Suppose a homogeneous and quiescent explosive atmosphere is prepared in the volume V. T is gradually increased. The combustion is active, but proceeds extremely slowly. If the heat losses (volumetric rate in W/m^3) are larger than the heat released by the combustion ($Q_{losses} > Q_{comb}$), the chemical reaction is dampened because T is forced to decrease. Because of the exponential temperature dependency of the reaction of combustion, there is a temperature where the heat released by the combustion becomes larger than the linear heat losses ($Q_{losses} < Q_{comb}$). Above this temperature nothing prevents the runaway of the reaction and the explosion occurs (T increases up to T_{ad}).

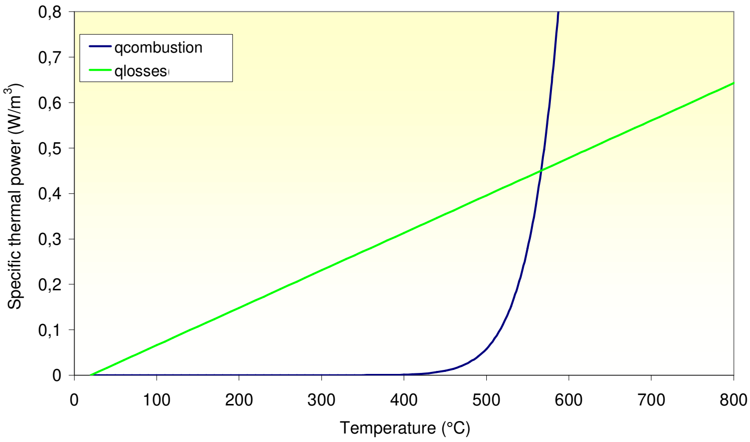

Fig. 1: *volumetric heat release rate and losses in a volume V (featuring the ASTM flash for autoignition temperature measurements) containing a homogeneous and quiescent stoichiometric hydrogen air mixture*

Although it is an over-simplified theory, it is useful to investigate the various ignition modes (Carleton and Weinberg, 1994). A well-known application, particularly fitted to this modelling concept, is the autoignition problem. The autoignition temperature of hydrogen mixture is about 550°C, measured in a 50 mm size flash. The temperature of the flash is progressively increased until the volumetric ignition. The latter occurs when the heat loss curve is tangent to the heat release curve[1]. This theory was applied in the present work to fit the combustion parameters (A and E_{act}/R). A good agreement (Fig. 2) is obtained with $A= 5.10^3\ s^{-1}$ and $E_{act}/R=10000$ K. Note this value for E_{act}/R is that recommended for hydrogen air mixtures (Coffee et al., 1983).

[1] Standard natural heat convection laws are used in [3]

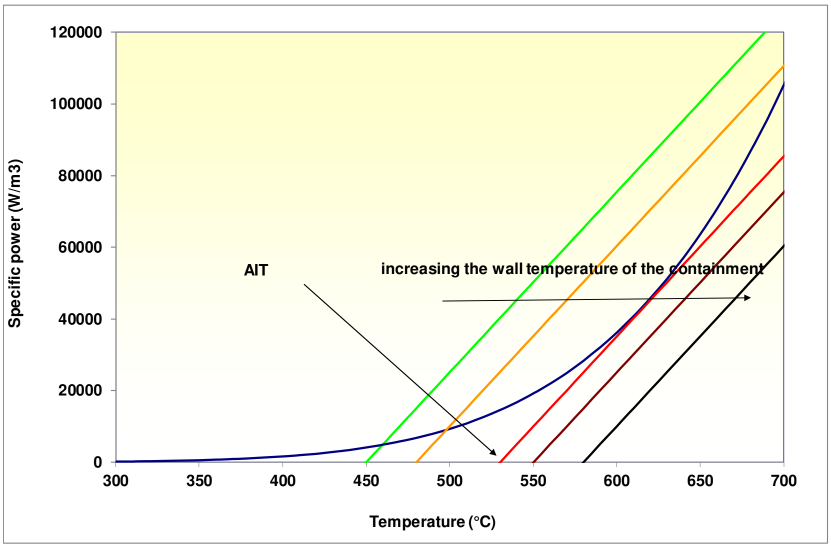

***Fig. 2 :** fitting of the combustion parameters to retrieve the autoignition temperature of hydrogen-air mixtures*

This modelling approach is now tentatively applied to spark ignition mechanisms.

In the spark gap, the heat of the ignition source, E_{spark} (in J), is delivered punctually and instantaneously. At this point, D=0 and T is infinite, so that heat losses are infinite and combustion cannot start. Some (small) amount of time is required for this initial "point" to spread out due to thermal conduction. Following, this initial amount of heat is spread by thermal conduction in the surrounding area but conserved if other thermal losses are ignored (heat radiation, thermal conduction to solid bodies). All along this "spreading" process", the spark energy is conserved according to the following expression (where D increases with time and Cp is the specific heat of the mixture in J/kg.K) which is an application of the first principle of the thermodynamics :

$$E_{spark} = \rho_0 \cdot Cp \cdot \frac{\pi}{6} \cdot D^3 \cdot (T - T_0) \quad [4]$$

The heat loss curve formulated by equation [3] (with h_{conv}~ 2.λ/D where λ is the thermal conductivity of the mixture at the spark temperature so approximately 0.1 W/mK) can now be computed using equation [4]. For each spark kernel size, a temperature and a specific heat loss can be calculated. This evolution is presented in fig. 3 for various spark energies (curves labelled "spark").

The heat release represented by expression [2] can now be plotted using the values of E_{act}/R and A found above. This curve stops at T=T_{ad} with is the maximum combustion temperature. When E_{spark} is too small (E_{spark}=0.015 mJ), the « spark heat loss » curve (line+triangles) never intercepts the heat release curve and, for any value of T, $Q_{losses}(T)>Q_{comb}(T)$ and the explosion cannot occur. When E_{spark} is sufficiently large (E_{spark}=0.05 mJ), the « spark heat loss » curve (line+circles) intercepts the heat release curve and the explosion can occur since there is a domain where $Q_{losses}(T)<Q_{comb}(T)$ while $T<T_{ad}$.

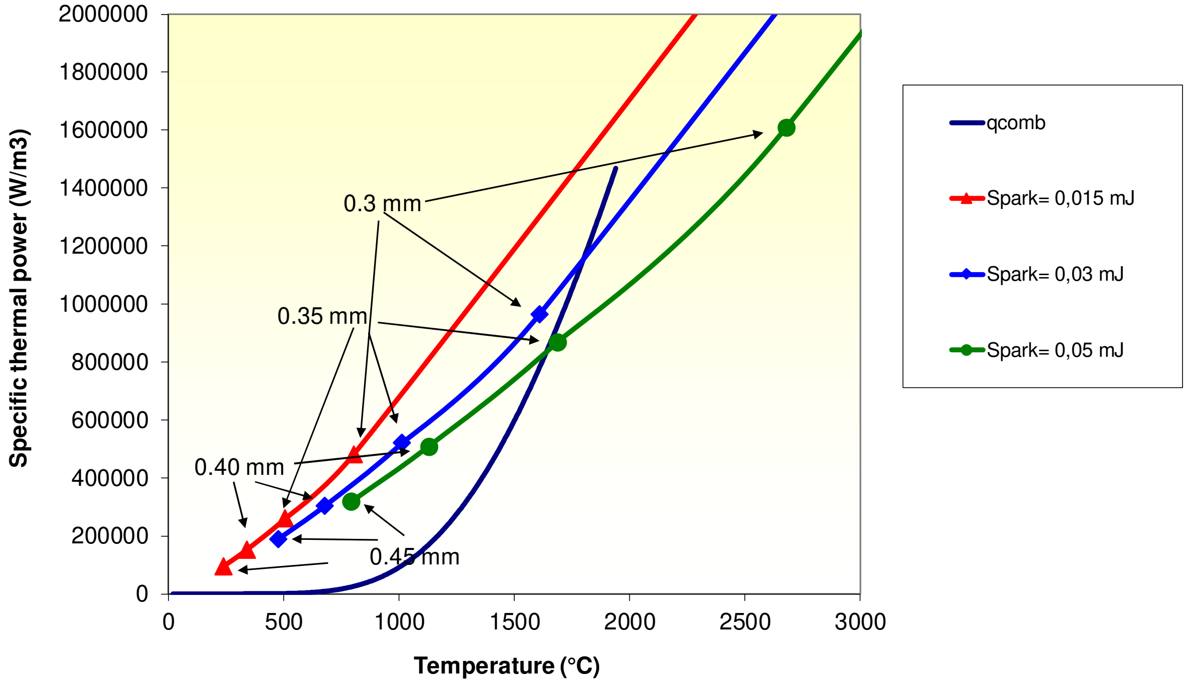

Fig. 3: *simulated spark ignition of a hydrogen-air stoichiometric mixture for three different values of the initial spark energy (arrows and text refer to the hot pocket of gas size at different times)*

The Minimum Ignition Energy (MIE) is thus obtained when the intercept is obtained for $T=T_{ad}$, corresponding to the upper point of the heat release curve. This particular point corresponds to a specific size of the hot pocket of gas called the « minimum flame kernel », D_{crit}, which is an intrinsic property of the mixture. It is the minimum size of a flame able to develop in a cold environment. This definition is very close to that of the « minimum quenching distance »[2] (D_{quench}). Physically, both parameters can only be proportional.

The main outcomes from this simplified theory are :

- In the MIE conditions (E_{spark}=MIE), the temperature of the spark kernel is equal to T_{ad} so that equation [4] can be used to calculate the MIE;
- T_{ad}, D_{crit} (D_{quench}) and MIE are interlinked. At the ignition point, [2] and [3] are equal and $T=T_{ad}$ and $D=D_{crit}\sim D_{quench}$. In equation [3], ($T_{ad}$-$T_0$) should be replaced by a parameter proportional to MIE/($\rho_0.\pi/6.Cp.D^3$) from equation [4] so that it appears that Q_{losses} is proportional to MIE/D^4. In addition, it can be shown that $\rho_0 \cdot A \cdot Cp \cdot (T_{ad} - T_0) \cdot \frac{T_0}{T_{ad}} \cdot e^{-\frac{E_{act}}{R \cdot T_{ad}}}$ is proportional to the square of the laminar burning velocity Sl_{ad} of the mixture (Glassmann, 1977). Further, it is known (Hattwig and Steen, 2004) that D_{quench} and Sl_{ad} are correlated:

$$Pe = \frac{Sl_{ad} \cdot D_{quench}}{a_{diff}} \approx 50 \qquad [5]$$

[2] the minimum quenching distance is the smallest gap between two cold parallel planes just allowing the flame to propagate steadily. If the distance is infinitesimally smaller, the flame is quenched.

where a_{diff} is the thermal diffusivity of the (cold) mixture (typically 2.10^{-5} m^2/s for ambient air). So, Q_{comb} is proportional to $1/D^2$. Since Q_{losses} is proportional to MIE/D^4, MIE should be proportional to the square of $D=D_{crit}\sim D_{quench}$.

This relationship was discussed Lewis and von Elbe (1987) and this last conclusion is fully confirmed by available data (fig. 4). MIE is approximately proportional to the square of D_{quench}.

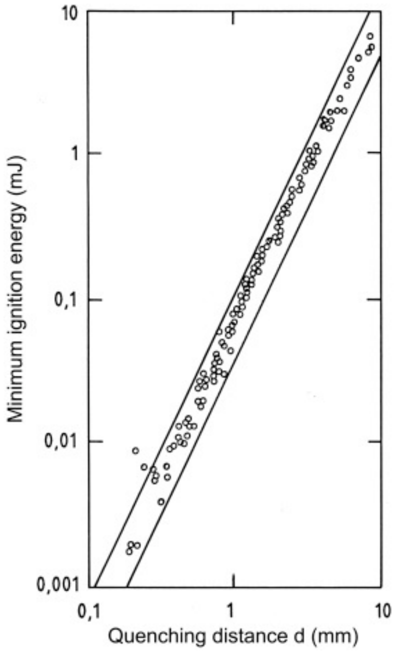

***Fig. 4:** relationship between MIE and D_{quench} (from Kuchta, 1985)*

The practical conclusions are that MIE is a fundamental parameter, depending only on the physico-chemical properties of the mixtures, and that it may be deduced from other fundamental parameters like D_{quench}. D_{quench} is not easy to measure and rather the Minimum Experimental Safety Gap (MESG) is used to correlate with MIE. But these are approximate correlations, relying on several assumption like the validity of [5]. Direct measurements of MIE are preferable when some accuracy is required.

2.2. Measuring the MIE

If a "spark" were truly "a punctual and instantaneous deposit of energy", the temperature would be infinite which is unrealistic. In practice, the deposit is not punctual nor instantaneous. Electrical deposits (breakdown, arc) or focusses laser beams are used in which plasma temperatures up to 5000 to 10000 K are reached in nanoseconds.

Usually, capacitive spark discharges are used because of the apparent simplicity (Kumamoto et al., 2011) since the energy delivered into the circuitry cannot be larger than that stored in the capacitor[3]. This energy can be easily varied, changing the charging voltage. Most often, the MIE is the minimum stored energy enabling ignition. A good overview was proposed by Ngo (2009). Usually, the capacitor

[3] Other techniques were tried like focussed pulsed laser beams (Weinrotter et al., 2005). The energy of the pulse is known but not the quantity really absorbed in the plasma created at the local point. The major part of it is transmitted through the plasma and dissipated by absorption in other parts of the beam. Following the minimum energy pulse (MPE) appears on order of magnitude larger than the traditional MIE.

is mounted in series with the spark gap and the voltage of the capacitor is increased slowly until breakdown. In some case, a transformer is used to produce the breakdown from a low voltage capacitor. A significant body of work was performed more than 50 years ago by Lewis and von Elbe (in the edition of 1987) and Calcote et al. (1952). The influences of several parameters like the spark gap distance and of the duration of the spark were investigated. They found that the optimum spark gap distance is about D_{quench}. When it is larger, the part of the energy of the spark dissipated outside the initial flame kernel is lost and when it is smaller the quenching effect of the electrodes appears. Because of this last phenomenon, the shape and diameter of the electrodes is also important especially when the spark gap is smaller than D_{quench}. It is advised to choose a spark gap slightly larger than D_{quench} and pointed and thin electrodes. Further, a shallow minimum as function of the duration of the spark is found. This optimum amounts a few tens of microseconds. Presumably for very short spark durations, the temperature of the plasma is so large that significant losses by heat radiation occur (and some mechanical losses by the emission of a shock wave). And obviously the energy delivered by the spark after the flame has emerged from the initial kernel is also lost.

Despite the effort devoted to controlling the spark, authors generally observe a large overlap between the “ignition points” and “no ignition” points (fig. 5). Because of this, experimentalist use some statistics, like probit laws, to define an ignition threshold (Eckhoff, 2010). Then, the renown ignition threshold, as those given by Lewis and von Elbe, might then be more a statistic limit than a true physically meaningful border (Bane et al., 2010). This statistical nature may come partly from the difficulty to master the spark characteristics although the flame ball development although the complicated flow produced in the spark may disturb the flame ball growth (Kono et al., 1992).

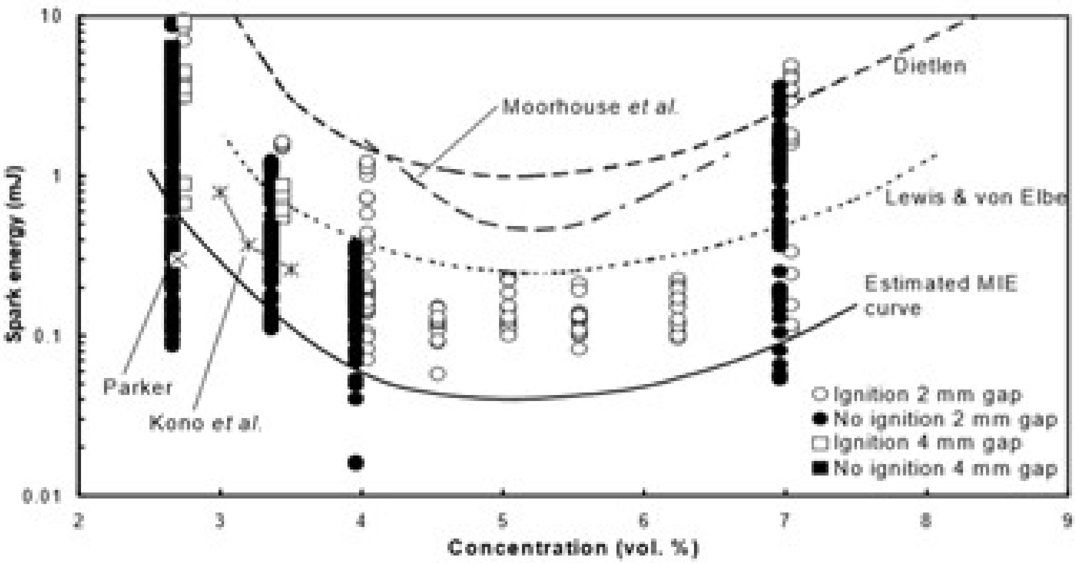

Fig. 5 : *illustration of the data scattering about the minimum ignition energy of propane-air mixtures at ambient conditions (from Randeberg and Eckhoff, 2007)*

It is believed that there is thus room to progress into the way of measuring the MIEs and, especially, in controlling the spark characteristics.

3. Experimental setup

The explosion chamber is a 7 liter transparent cylindrical chamber (figure 8) able to support a static internal pressure amounting 100 bars. It is equipped with measuring ports and feeding lines. The mixture is prepared into a small stainless-steel mixing tank (0.1 l) using mass flowmeters (fig. 6). The composition is controlled using a paramagnetic oxygen meter (SERVOMEX : O_2 % accuracy = ±0.01 % v/v) upstream of the chamber and downstream. For a targeted 5% v/v fuel/air mixture, the accuracy of the composition is : 5±0.1 % v/v. The maximum explosion (over)pressures is measured using a Kistler 10 b piezoresistive gauge. High speed video (up to 10000 fps: Photron camera) is used.

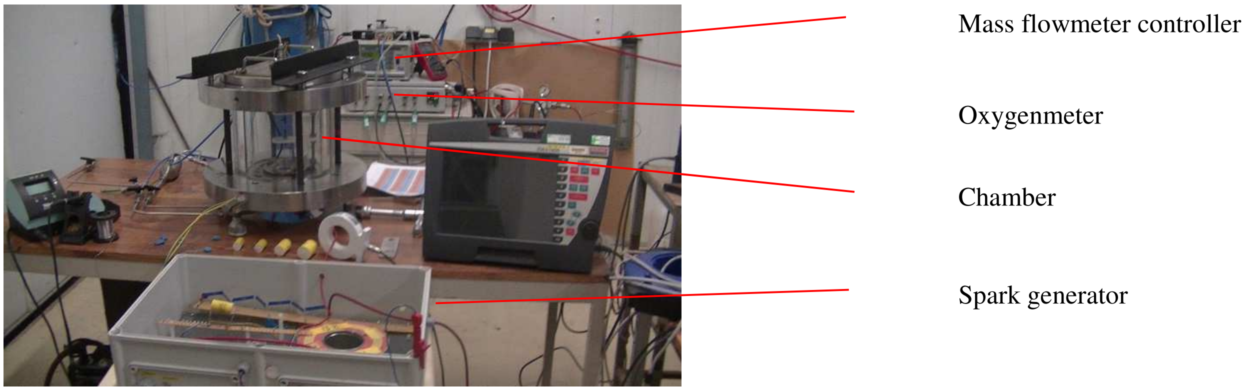

***Fig. 6 :** experimental set-up*

In the spark gap a part of the electrical energy is dissipated into heat which is the source of the ignition. But part of it is lost in producing the conditions sustaining the current stream (vaporizing the material of the electrodes, anode/cathode voltage gap,..), heat conduction towards the electrodes, shock waves, thermal radiation,… Microcalorimetric measurements (not shown here) showed that using small tungsten electrodes is a good mean for reducing the first two sources of losses. The electrodes are 0.3 mm in diameter (Tungsten) tightly screwed on steel rods in such a way that any heat loss sources (supports, walls) are cms away from the spark gap (fig. 7).

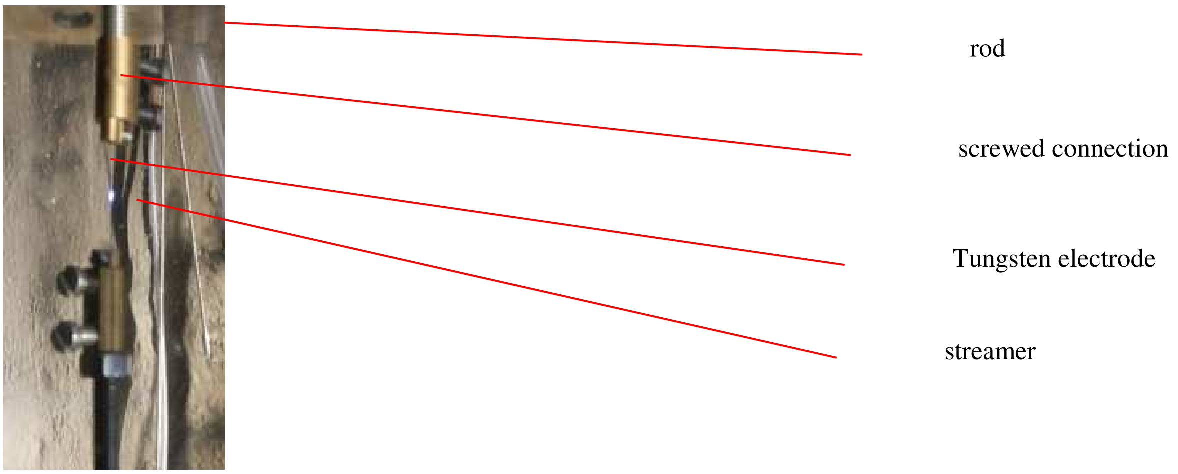

***Fig. 7** : spark gap arrangement*

But only a better control of the current and duration of the spark can reduce the two last sources of losses which supposes a flexible control of the current/voltage supply which depends on the spark generator. A large variety is described in the literature. An electrical spark is a two steps process. During the "disruption phase", a large voltage is required (typically 3000 V/mm) to ionize the atmosphere and create a "streamer" (ionized channel visible on fig. 7). The current remains very low during that phase (micro amperes). Then the electrical arc takes place during which a large current (up to tens of amperes) flows through the spark gap. The voltage amounts a few volts to several tens of volts. As said before, in many spark generators, the electrical energy is stored in a single capacitor and is used to create the disruption and the arc with considerable losses due to very large currents. The ratio energy delivered in the spark/stored energy may vary is significant proportions depending on the current, so from one test to another. This might be a reason for the statistical nature of the ignition. To obtain a better yield, the impedance of the circuit needs to be carefully controlled. This was attempted in the present work.

If a purely capacitive circuit were used with only a few meters of cables, the expected resistance would be a fraction of 1 ohm and the inductance nearly zero. Using a single high-voltage capacitor and a 2 mm spark gap, a minimum voltage of 6000 V would be required meaning a typical capacitor value of 5 pF (this is difficult to control in practice) to be able to deliver 100 µJ. The energy is dissipated into the circuit within a time scale amounting R.C= 10^{-12} s which is much too short for an efficient ignition and with a very large current (hundreds of amperes) favoring the losses. This is the reason why it is suggested, in the standards for instance, to add a rather large inductance (1 mH). Its role is to increase the impedance of the circuit without consuming much energy (which a resistor would do) to reduce the current and increase the duration of the spark. However, our initial trials showed that controlling a very low capacitance circuit is difficult and that the scattering in the disruption appearance is significant depending presumably on the electrodes (shape, surface conditions,…). This technique was rapidly abandoned.

Rather, it was decided to disconnect the two phases of the spark. A separate high voltage and high impedance circuit is used to produce the streamer. A second, low voltage and low impedance circuit, is used to produce the arc and dissipate the energy. Once the streamer is created the low voltage circuit discharges automatically. A set of Zener diodes prevents the high voltage to be transmitted to the low voltage circuit. The circuit is presented on fig. 8. The current is measured using a standard current gauge and the voltage is measured at the charge capacitor. As it stands, the total capacitance of the circuit is 20 nF (which is that of the charge capacitor but which can be varied), the inductance is 1.7 µH and the resistance is 0.1 Ω. The measured capacitance of the chamber, of the diodes and of the high voltage cable is not more than 45 pF, half of this being due to the cable. An additional 1 mH inductance can be placed close to the electrodes. The main advantage of the device is that the stored energy can be varied in very large proportions since it was verified that an arc is maintained down to voltages as low as 35 V. In theory, the stored energy can be varied between 10 and 2000 µJ without changing the capacitor. Note that it was verified that the streamer is not able to ignite the mixtures.

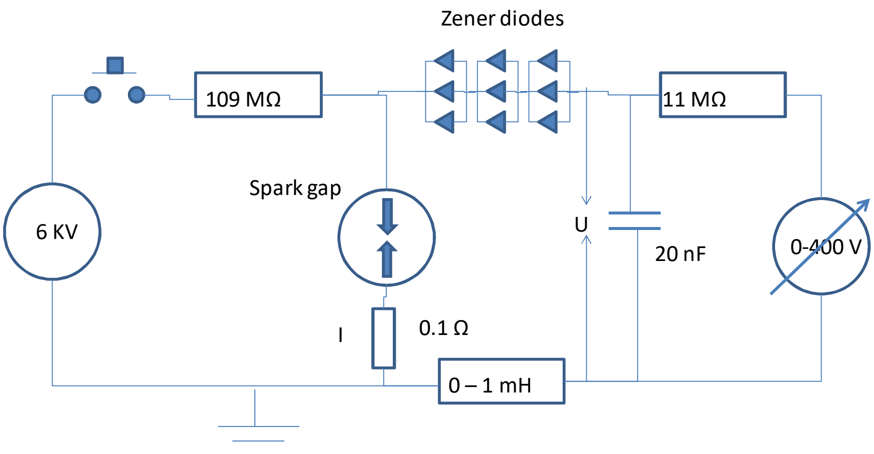

***Fig. 8 :** scheme of the electrical circuit*

Electrical tests were performed without the high voltage part (to limit the noise affecting the very beginning of the signal at the start of the breakdown). In the first test (fig. 9), the diodes were short-circuited and the discharge was obtained by short circuiting manually the electrodes. Doing this, a short spark is produced over a length amounting a fraction of a mm during some tens of microseconds.

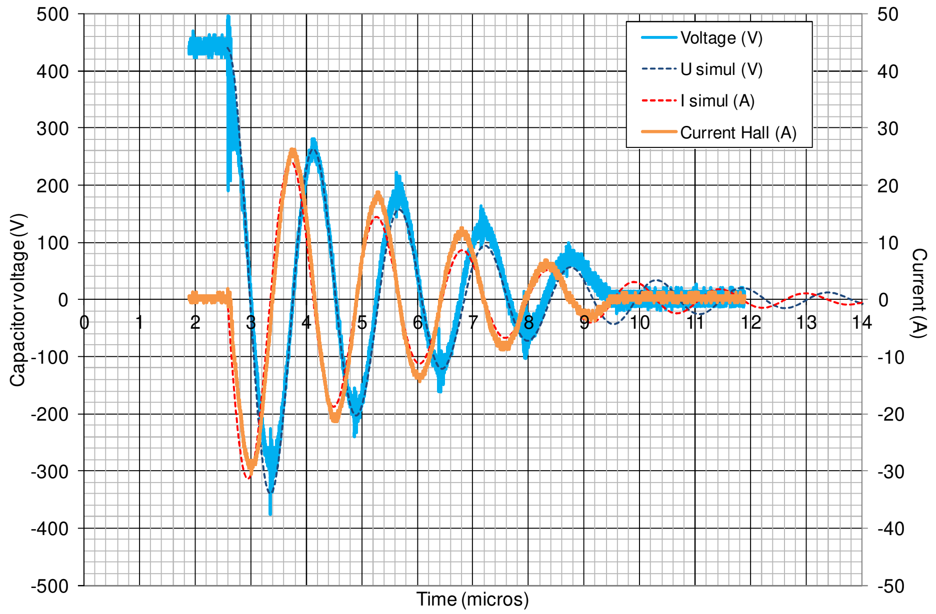

Fig. 9 : *current voltage signals : no streamer, manually triggered spark, without additional inductance, 440 V in the 20 nF capacitor, without the diodes.*

In the second test, the diodes were reintroduced (fig. 10).

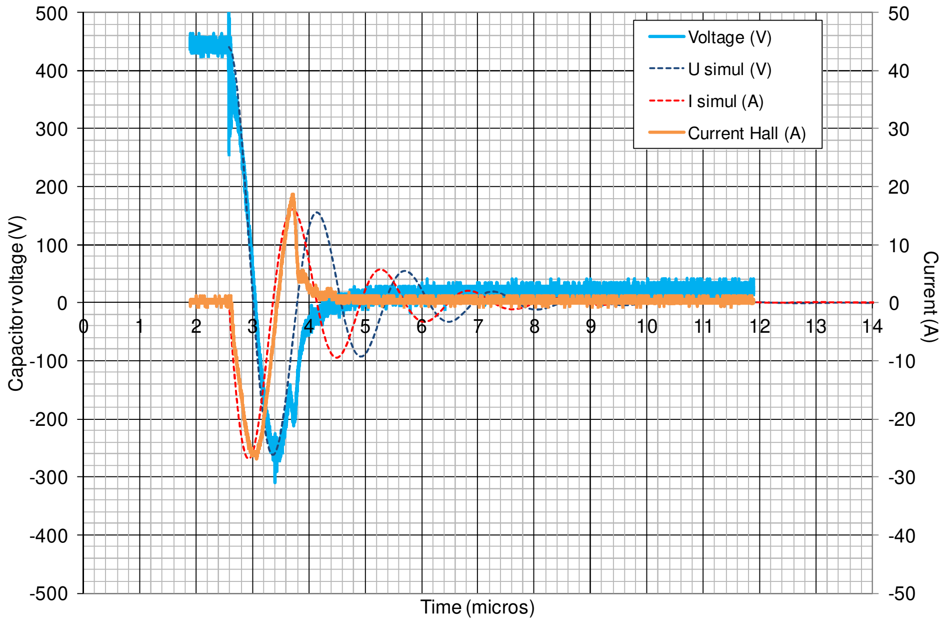

Fig. 10 : *current voltage signals : no streamer, manually triggered spark, without additional inductance, 440 V in the 20 nF capacitor, with the diodes.*

The dampened oscillating signal of figure 9 is a typical RLC circuit response with a moderate resistance. Clearly, the diodes have a strong influence (fig. 10) and remove a lot of energy. A simulation of the RLC circuit was performed, and the results are the dotted lines on fig. 9 and 10. The best fit is obtained with an inductance of 3 µH (capacitor=20 nF) and a resistance of 2 Ω for fig. 9

and an inductance of 3 µH (capacitor=20 nF) and a resistance of 4 Ω for fig. 10.When the diodes are short circuited, it can be verified that all the energy is consumed in the resistance. The latter is much larger than the natural resistance of the wires as measured initially. Most probably it is that of the "spark gap" (the direct contact of the electrodes). When the diodes are incorporated, the best fit is obtained with R=4Ω at least until about 3.5 µs. After this, a catastrophic change appears suggesting a strong increase of the resistance as the Zener diodes start to open the circuit. The characteristic of the circuit turns from a LC to a RC dominated. In terms of energy, the average resistance needed to close the balance would be 6 Ω. About half of the energy is dissipated in the "spark gap" and the duration is on the order of 1 microsecond.

A similar attempt was performed inserting a 1 mH inductance (added resistance=2Ω). The arc is triggered by the streamer (2 mm spark gap). The signals (fig. 11) resembles that of fig. 10 but over a larger timescale (10 µs instead of 1) as expected. The very beginning of the current signal is an artifact probably due to the Zener diodes closing the low voltage part of the circuit. The simulation shows that the best fit is obtained with L=0.4 mH (instead of 1 mH) and R=150 Ω. This value of the resistance closes the energy balance as well. This increase of the resistance as compared to figure 14 is due the spark gap which resistance increases in proportion with the electrode distance and inversely with the current intensity. From the energy remaining in the charge capacitor, current measured and model of the circuit it was possible to obtain a robust estimate of the yield of the spark. With a reduced current below 1 A, the yield is better, up to 90%. With a current of about 3 A, the yield is 70% at 10 A, it drops at about 50%. To some extent, it confirms that depending on the voltage between the electrodes when the spark appears in standard apparatuses, the yield of the spark may strongly vary.

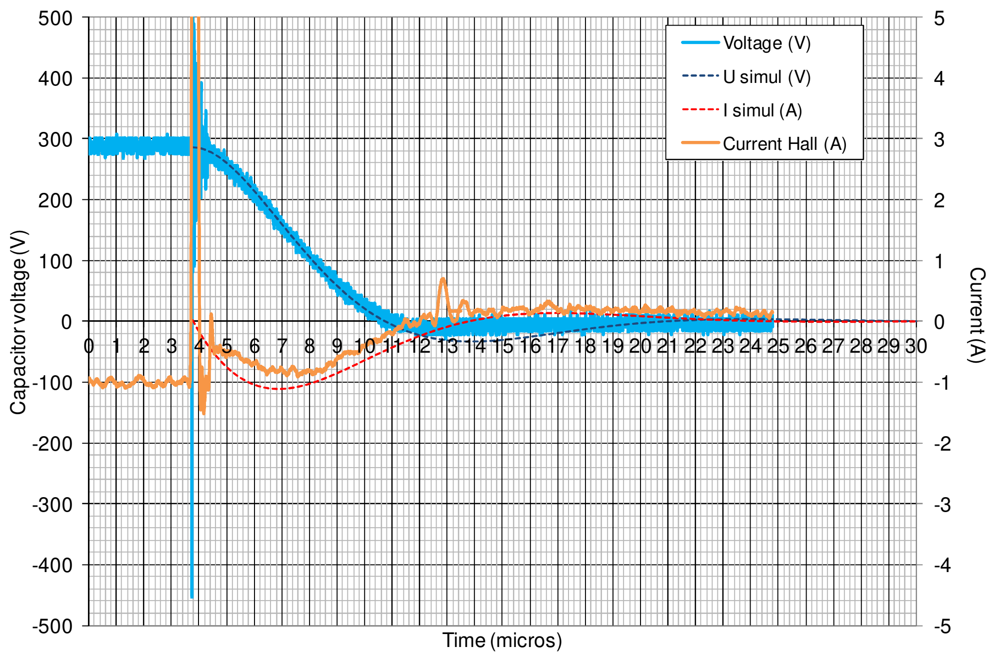

Fig. 11 : *current voltage signals : streamer triggered spark, with a 1 mH additional inductance, 290 V in the 20 nF capacitor, with the diodes.*

4. Results

Propane-air and hydrogen-air tests were performed.

A typical sequence is shown on fig. 12 for a 4.5% v/v propane-air mixtures. The spark gap is 2 mm. A double lobed structure appear just after the spark with seems to be a double vortex and would suggest a Maecker (1955) effect due to the flow of heavy ions between the electrode. This "jet" is mostly responsible for the energy transfer between the spark gap and the mixture. A blue flame kernel is visible at 3 ms and expands out. The expansion velocity is a few m/s.

The minimum ignition energy measurements are shown on fig. 13a (the values are three quarters the energy stored in the capacitor to account for the losses). The voltage is dropped gradually until systematic no ignition. Each test is repeated 10 times. There is some remaining overlap between the no ignition zone and the ignition zone but much less at compared to the data of fig. 5. It is possible that the remaining scattering results from the development of the flame kernel (Esmann et al., 2020). If the minimum ignition energy is in line with the standard values (0.3 mJ at stoichiometry fig. 13b), the evolution of MIE on the lean and rich side are different but in line with the most recent measurements.

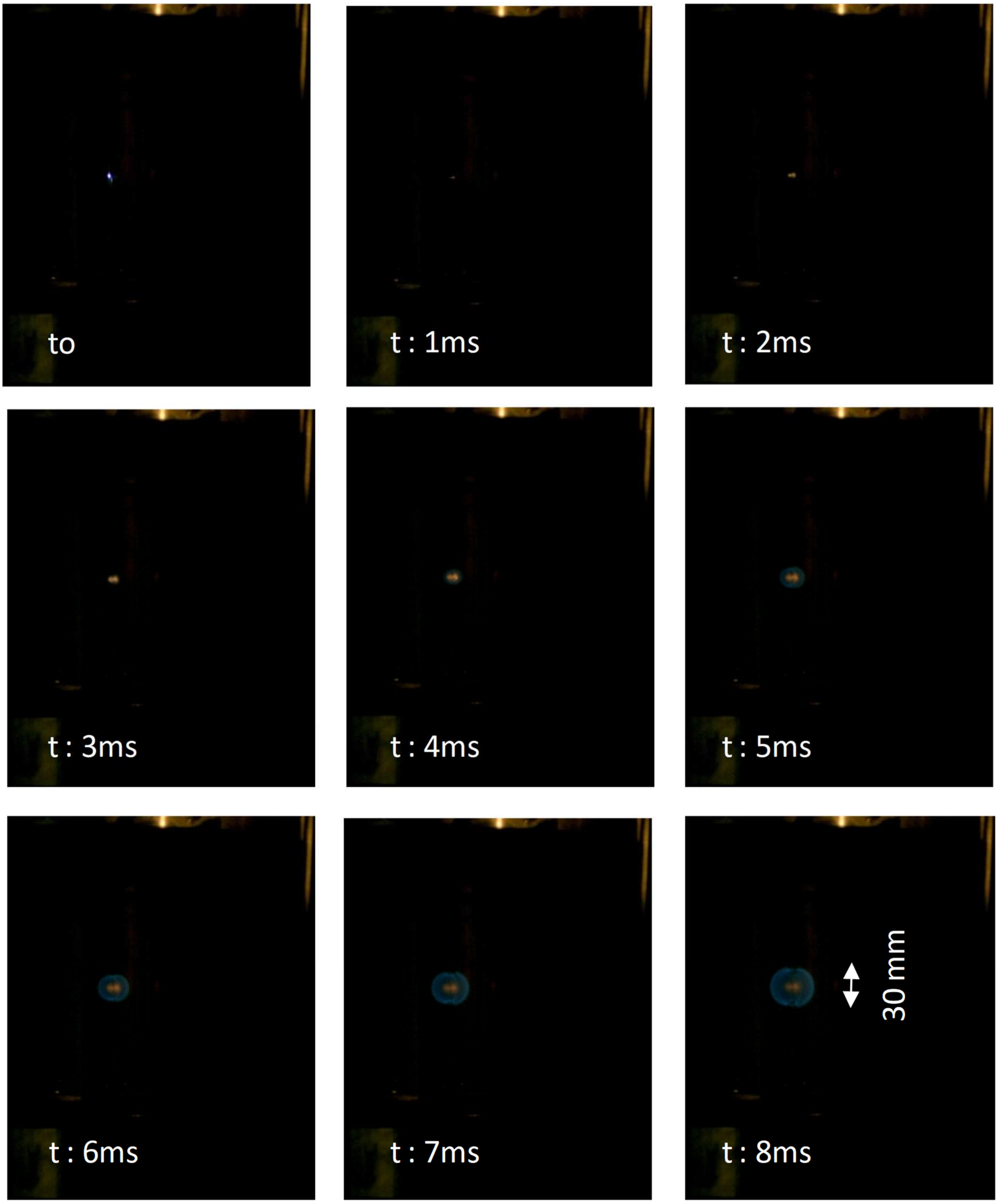

***Fig.12 :** flame kernel development (4.5% C_3H_8 in air, 400V, 1 mH)*

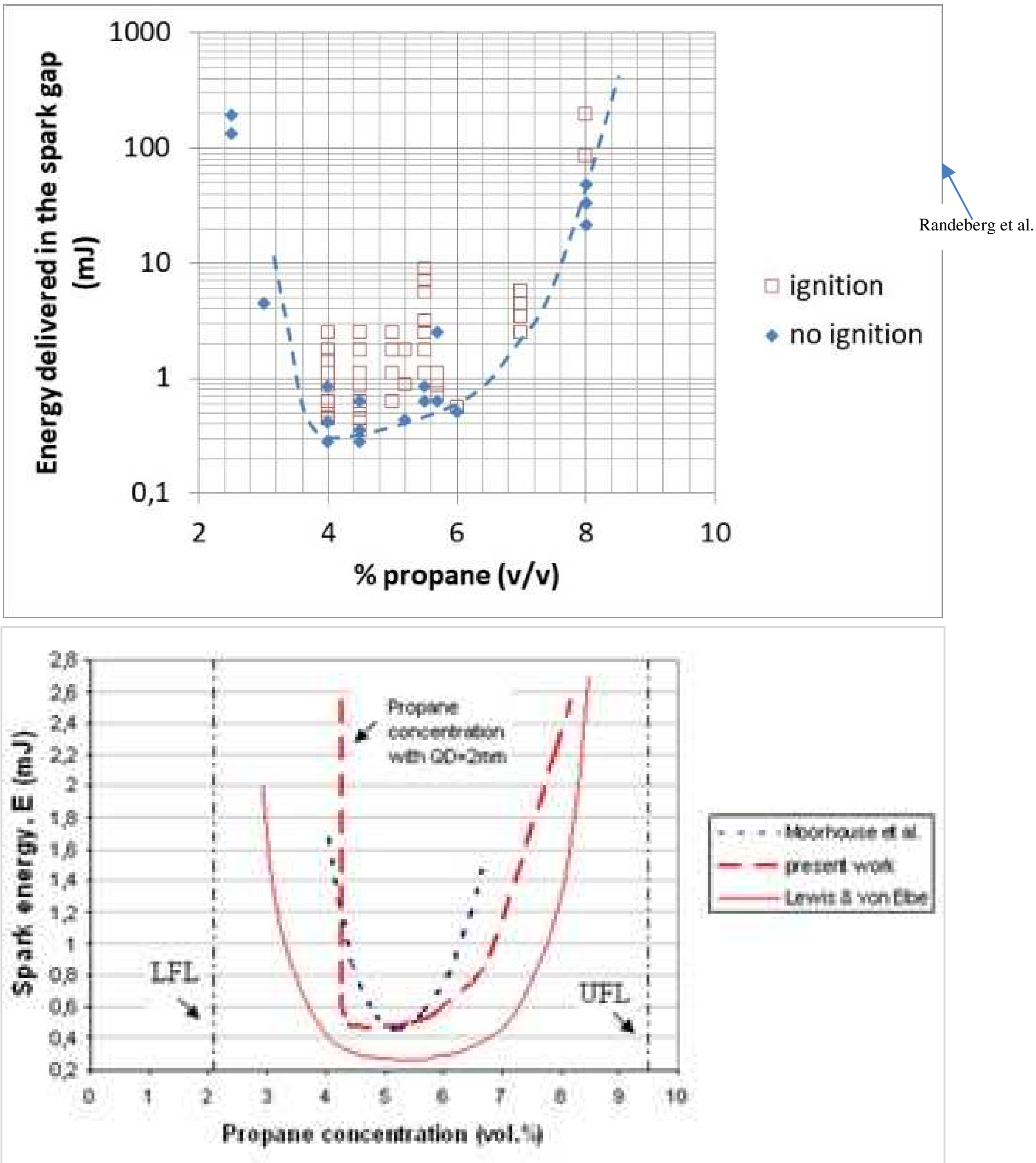

***Fig. 13 :** minimum ignition energies for propane air mixtures at ambient conditions (bottom grap^h from Randeberg et al., 2007)*

A typical sequence is shown on fig. 14 for a 40% v/v hydrogen-air mixtures. The spark gap is 0.5 mm. the beahaviour of the flame is similar to that of the propane air mixture.

The results of the MIE measurements for hydrogen-air mixtures are shown on fig. 15. In fig.15a are given the results obtained under exactly the same conditions than for propane-air experiments. In these tests the mixture is still. In fig15b, the same measurement obtained with a continuous flow of the flammable mixture (0.1 m/s). There is a clear influence of the experimental conditions. It was noticed during these tests the extreme sensitivity of the triggering of the spark to the presence of humidity. It is believed that when the mixture is still, the water vapor produced by the local burning of hydrogen modifies the sparking conditions especially at the easiest burning conditions (40% H_2 v/v) for which the sparks are very tiny. The present results are globally consistent with the literature (Ono et al., 2007).

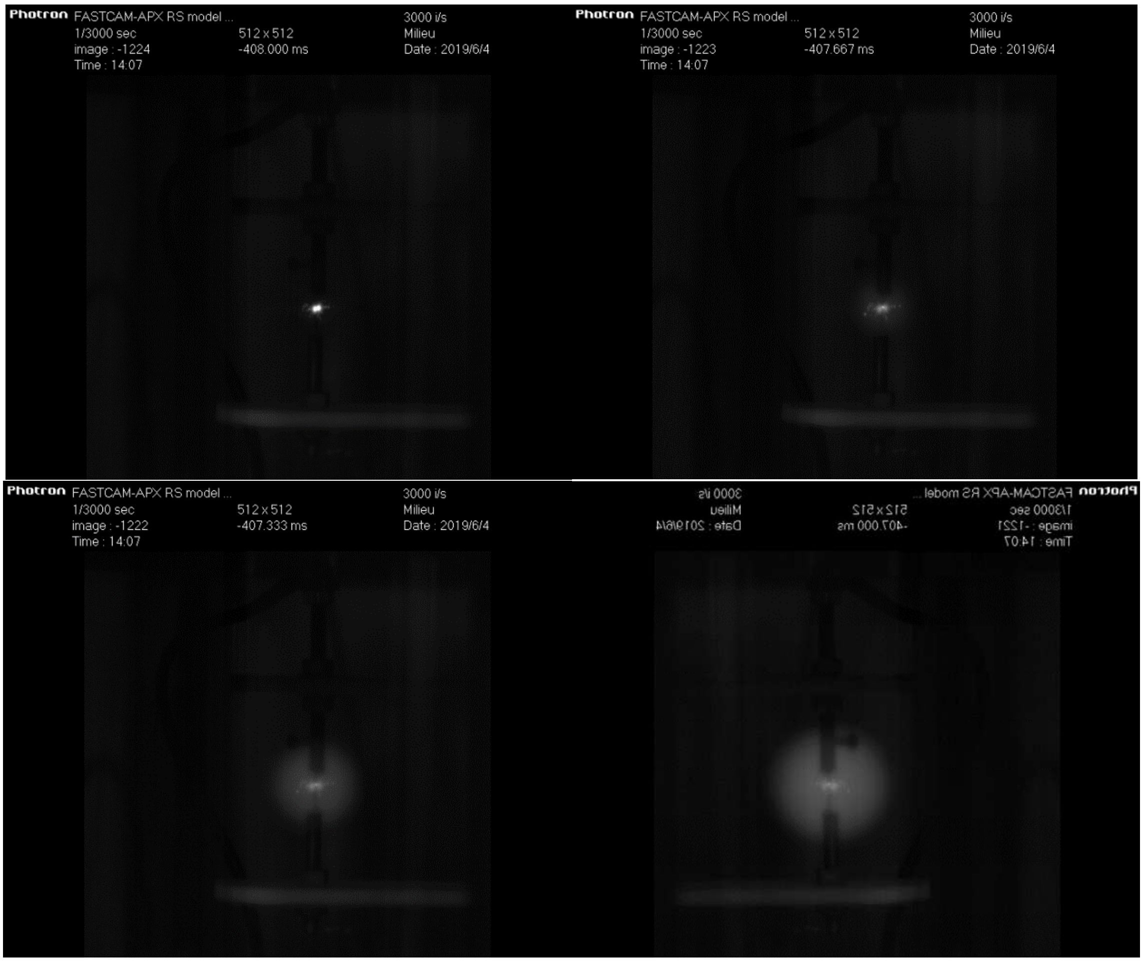

***Fig. 14 :** flame kernel development (40% H_2 in air)*

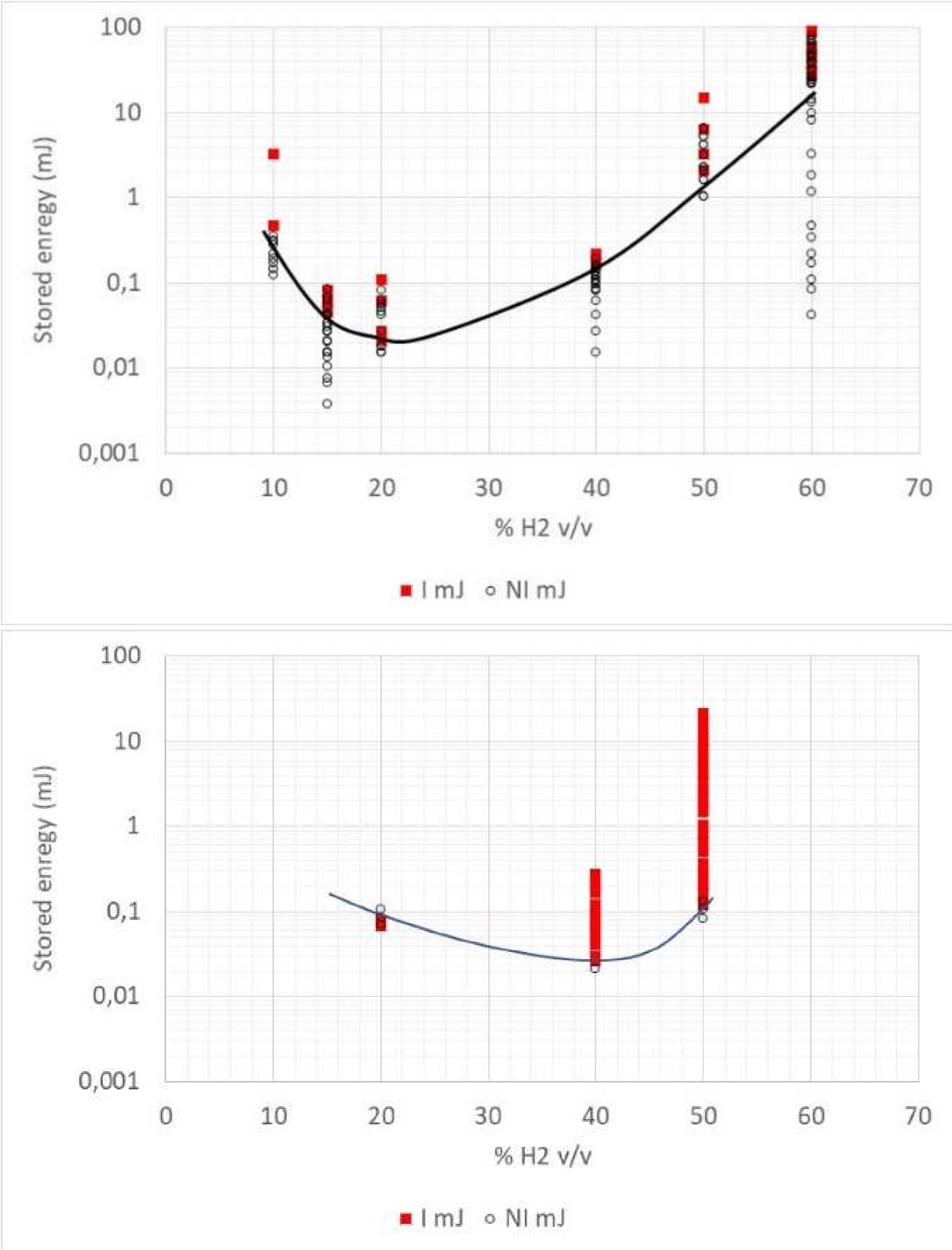

***Fig. 15 :** minimum ignition energies for hydrogen air mixtures at ambient conditions (a-with a still mixture and b- with a flowing one)*

5. Conclusions

In the present document, a new device to measure minimum ignition energies of flammable gases is presented. The originality of the system lies in the triggering mechanism of the spark which promotes the reproducibility by controlling the losses.

It is shown that the scattering of the ignition/non ignition point is can be reduced as compared to other studies. However, for very low MIEs, like with hydrogen air mixtures, the ignition process seems more erratic although the sparks are reproducible. Humidity seems to be a potential explanation.

Acknowledgements

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