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Ignition of Lean Air / Hydrocarbon Mixtures at Low Temperature by a Single Corona Discharge Nanosecond Pulse

A great number of experimental studies have demonstrated that non-thermal plasmas produced by high voltage pulse discharges, running at a given pulse repetition frequency, are able to ignite air / hydrocarbon mixtures at a low initial temperature and atmospheric pressure. In this paper, we show that ignition can also be achieved using a single nanosecond pulse corona discharge generated under a very strong overvoltage. Experiments were conducted in air / propane and air / n-heptane mixtures. For such a discharge, ignition of n-heptane requires less released electrical energy than propane and lean mixtures can be completely burnt with reasonable energy values of several tens of mJ. Extended flame kernels or several points of ignition can be induced. For n-heptane, we have developed a simplified combustion model using a reduced mechanism, enabling us to suggest a kinetic explanation for ignition at a low temperature. In particular, it appears that oxygen atoms produced in the plasma induce a significant reduction of the ignition time. This model lays the foundation for a more complete study, including the production of other active species by the discharge, such as the first excited state of the oxygen atom, or even dissociation products of the hydrocarbon molecule following electron collisions or quenching of the nitrogen excited states.

Introduction

Combustion control and ignition using cold and non-thermal plasmas has become a major topic of interest over the past ten years or so [1, 28-31], and more specifically the use of discharges generated under strong overvoltages for automobile atmospheric engine applications [7, 24, 36]. Due to strong environmental constraints for automobile exhaust gases, engines to be developed in the future must run on lean air / gasoline mixtures, or mixtures diluted with burnt gases. In both cases, the optimization of ignition devices is required, since classical spark gaps become inefficient under these conditions. In this context, the generation of non-equilibrium plasmas in large volumes, with high densities of active species, and the ability to induce rapid gas heating is challenging. Among the various types of electrical discharges studied, the pulsed corona generated under a very strong overvoltage has recently demonstrated promising characteristics for ignition issues at low initial temperatures (close to 300 K) of air-based gas mixtures at high pressure (at least one atmosphere) [3, 16, 33, 34]. This discharge should also be useful for the ignition of mixtures with a reduced oxygen concentration (less than 20%) at a pressure lower than one atmosphere and thus may be interesting for aeronautical applications. Another concept is the use of a discharge to produce excited molecular oxygen species to be then injected into the combustible mixture [27].

It was shown that the plasma produced by a single nanosecond pulsed overvoltage in a point-to-plane configuration can be used to ignite various kinds of quiescent air / fuel (propane C_3H_8, n-heptane n-C_7H_{16}) mixtures [3, 16, 33]. Diffuse and large plasma is able to develop in atmospheric air [34, 35]. This diffuse pattern is created by a multi-avalanche overlapping process. Space charge mechanisms and screening effects responsible for streamer inception and multi-channel generation are weaker, since the applied electric field increases very fast over the entire gap. The addition of propane into air induces a significant effect of diffuse discharge constriction very near the anode and its filamentation within the other part of the gap [3, 4, 16]. However, the discharge is able to ignite mixtures along paths between the two electrodes and not only at the pin, which leads to a specific cylinder-shaped flame propagation instead of a classical spherical one [5, 6, 16].

Several results emphasize the possible advantages of such an igniter, compared to classical spark gap devices. First, very lean air / propane and air / n-heptane mixtures can be completely burnt with reasonable released electrical energies of several tens of mJ. Sparks remain inefficient at very low equivalence ratios. Second, nanosecond discharges can induce extended flame kernels or several points of ignition. Spark gap ignition will always remain much localized at a single point. The consequences are faster combustion and less misfiring. Third, ignition times are reduced with non-equilibrium discharges. A thermal spark promotes neutral heating by maintaining the electrical current over a long period, but it is less efficient in the end. For example, T. Shiraishi et al [24] compared the ignition of iso-octane by a streamer discharge and by a thermal arc, and showed that the use of the non-thermal plasma is more efficient, in particular with a shorter ignition time, than the arc, whereas the arc current is maintained for a greater amount of time.

Decomposition mechanisms of hydrocarbons in the plasma phase deserve to be studied in more detail. In particular, data regarding the production of radicals or molecules (cross-sections and reaction coefficients) following dissociative excitations by electronic collisions or by quenching of the nitrogen excited states are rather scarce. The data proposed by other scientific communities must be clearly validated using dedicated experiments and by comparison with detailed kinetic modeling. Kinetic schemes developed for cold combustion should be taken into account in the interpretation of experimental results, including the effect of peroxy radicals, which are efficient combustion chain propagators through the generation of the hydroxyl radical. Experimental and numerical studies should be extended to heavy hydrocarbons, in order to simulate actual fuels, for example n-heptane or iso-octane (gasoline engines), aromatics and saturated monocyclic hydrocarbons (kerosene in aeronautical applications).

The mechanisms involved in the combustion of propane have been the subject of numerous studies [8, 10, 11, 26]. However, few things are known about combustion control and ignition of air / propane mixtures by non-thermal plasmas [12, 13, 18, 22]. Evidently, oxidation reactions of hydrocarbon molecules by oxygen atoms and by hydroxyl radicals should play a role in the ignition process, but such reactions are not so efficient at low temperatures for saturated compounds. Obviously a complete understanding of the ignition phenomenon requires a clear knowledge of the kinetic decomposition mechanisms of the hydrocarbon molecules in the plasma [19], as well as the production processes of the hydroxyl radical, which is more reactive with hydrocarbons than the oxygen atom is at a low temperature. Under conditions of a uniform plasma and electric field amplitude similar to that created during the diffuse corona discharge, a recent kinetic modeling by E. Filimonova [9] shows that the discharge stimulates the development and increases the intensity of a cool flame in an air / propane mixture. It sharply reduces the cool flame ignition time, as well as the total induction time. The rapid formation of propyl-peroxy radicals $C_3H_7O_2$ and propyl hydro-peroxide C_3H_7OOH in large concentrations is promoted, leading to a partial release of chemical enthalpy due to the organic peroxide decomposition. As a result, the discharge promotes faster oxidation of the mixture at initial temperatures in the range 500-800 K to the point of hot ignition occurrence (typically T > 1000 K). In other words, an air / fuel mixture is heated within the range of temperatures where the high temperature oxidation mechanism occurs with a higher rate compared to that of the auto-ignition case [9].

Studies using heavy fuels instead of small hydrocarbons (C1–C5) are not extensive [24, 25]. Recently, S. Nagaraja et al [17] numerically investigated the effect of a non-equilibrium plasma produced by a dielectric barrier discharge running at a high HV-pulse repetition frequency (60 kHz) on the ignition properties of n-heptane diluted in dry air at a reduced total pressure of 0.2 atm and with an equivalence ratio and initial temperature in the range of 0.5-1.5 and 550-650 K respectively. For this molecule, low-temperature chemistry plays a critical role. It is shown that O, H atoms and OH radicals produced by the plasma initiate and accelerate the H abstraction of fuel molecules (RH) and reduce the induction time of the exothermic cycle RH → R → peroxy RO_2 → OROOH by a factor of ten [17].

This paper gives, in a first part, an overview of experimental results obtained in our laboratory on the ignition of air / propane and air / n-heptane mixtures at a low temperature (ambient, close to 300 K) and atmospheric pressure, by a single corona discharge nanosecond pulse. In a second part, we present the results of a 0D-kinetic modeling, giving insight about the role of the oxygen molecule dissociation with regard to the ignition time in air / n-heptane. Such a model helps in the understanding of the ignition processes involved in corona discharges under strong overvoltages.

The nanosecond corona discharge and ignition of air / fuel mixtures

The experimental set-up and the associated diagnostics have been described in detail in previous publications [3, 4, 16, 33-35]. A positive high voltage (up to 50 kV) is applied between a parabolic pin electrode and a grounded plane over a single nanosecond range pulse with a short rise time of 2 ns. Changing the distance between the electrodes (within the range of 1-2 cm) and changing the HV-pulse length (10-60 ns) enables the electrical energy released into the gas (10-120 mJ) to be changed. This energy is determined by time integration of the electrical power given by the current-voltage product. The technique used for measuring the discharge current and voltage has been given by P. Tardiveau et al [34]. The plane is grounded through a 0.2 Ohm low inductance resistive shunt. Precise and time resolved current measurements can be performed with this coaxial type shunt without any electromagnetic disturbance. On the other hand, the voltage is recorded at the end of the HV-generator line using a home-made capacitive coaxial probe. This probe is completely integrated into the line, in order to have a high bandwidth and to not disturb the discharge. Signals are recorded on a 500 MHz bandwidth digital oscilloscope with a 4 Gs s^{-1} sampling frequency per channel.

The intense electric field (higher than 1 kV/cm near the tip, corresponding to a reduced field E/N greater than 400 Td, N being the total molecule density at 1 bar and 20°C) induced within the electrode gap promotes the development of a particular regime of atmospheric pure air discharge [34, 35]: it spreads over the entire electrode gap in a completely diffuse way, leading to a large volume of plasma. This diffuse regime tends to disappear in mixtures containing low hydrocarbon (HC) percentages and a multi-channel pattern develops, as shown in Figure 1 [3, 4, 16].
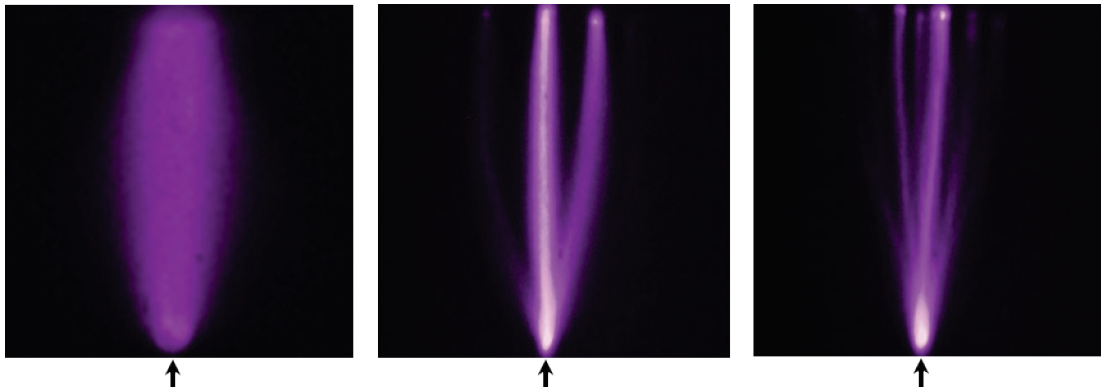
Figure 1 - Diffuse nanosecond discharge in pure air (left) and filamentation effect in air / propane (middle) and air / n-heptane (right) mixtures with 6 % of hydrocarbon. The arrow gives the location of the pin HV electrode.

We define the ignition threshold energy as the minimum electrical energy that the discharge must release into the mixture in order to ignite it and to induce flame propagation and complete combustion of the entire volume. Figure 2 shows how this minimum changes with the HC concentration. For stoichiometric mixtures (equivalence ratios equal to 1, for 4% of propane or for 1.9 % of heptane), this is around 17 mJ whatever the kind of hydrocarbon. It increases with lower equivalence ratios and this effect is much more significant for propane. The ignition of lean mixtures remains possible down to the low flammability level (2.1% for propane, 1.1% for n-heptane), but complete combustion cannot be achieved below 2.4% of propane, whatever the energy of the discharge. It is clear that ignition of the lean air / n-heptane mixture is much easier than for the air / propane one.
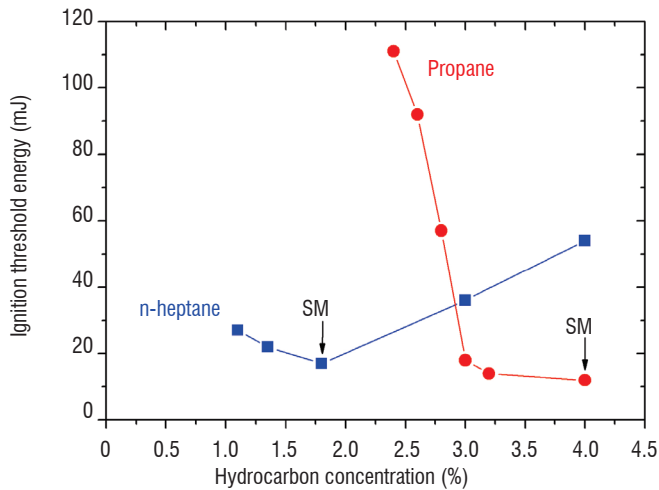

Figure 2 - Minimum energy for ignition as a function of propane and n-heptane concentrations (HV-pulse amplitude: 45 kV, HV-pulse length: 13 ns, electrode gap: 1.6 cm). SM: Stoichiometric mixture. Initial temperature: 300 K.

For low discharge energy (about 10 mJ) or low equivalence ratios, there is a single point ignition located at the tip of the anode and it leads to a classical spherical flame. However, in the case of a stoichiometric air / propane mixture, the increase of the energy up to 80 mJ leads to ignition all along the plasma channel. This can be identified in Figure 3A by the blue light in the first picture. If the energy density is high enough, the discharge can induce a kind of cylindrical flame and, in the first two pictures of Figure 3A, the cylindrical kernel is overlapped by two brighter flame kernels, starting at each electrode and leading to a rather complex flame structure. In the case of a stoichiometric air / n-heptane mixture, this cylindrical shape is no longer observed and only the two-point ignition remains, as shown in

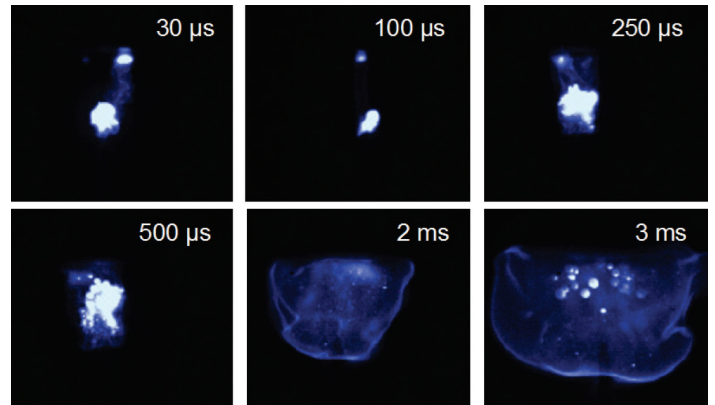Figure 3B. Two quasi-spherical kernels grow from the electrodes and merge together.

Figure 3a - Ignition along a plasma channel and cylindrical propagation in a stoichiometric air / propane mixture (Electrode gap: 1.6 cm, discharge energy: 85 mJ). Initial temperature: 300 K.
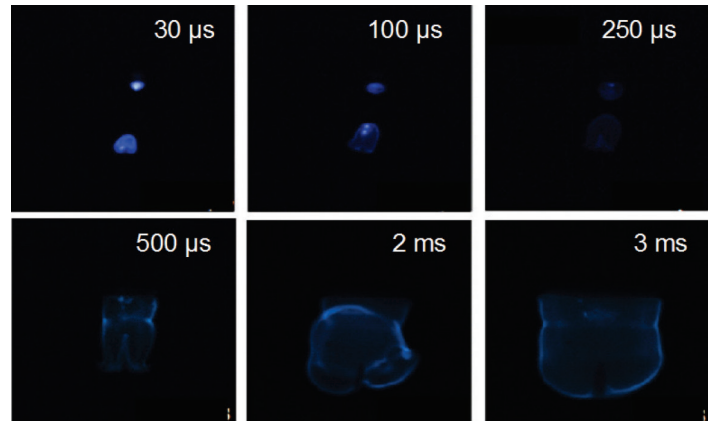

Figure 3b - Ignition along a plasma channel and cylindrical propagation in a stoichiometric air / n-heptane mixture (Electrode gap: 1.1 cm, discharge energy: 80 mJ). Initial temperature: 300 K.

The flame front speed is not dependent on the amount of electrical energy released into the gas and is only governed by combustion kinetics. However, the time needed to burn the whole amount of gas is dependent on this amount and, more precisely, on the local energy density. If the density is high enough over a long distance, as observed in propane at high energies, the initial flame kernel becomes longer and globally increases the rate of combustion. For stoichiometric conditions, the times required to achieve complete combustion can be reduced by twice as much as those obtained with spherical flames. This is also true for the double point ignition in heptane.

In the case of single point ignition, the times required for complete combustion of the gas mixture enclosed in the discharge chamber are given in Figure 4. Complete combustion is achieved more rapidly with n-heptane than with propane. This is due to higher combustion temperatures (2200 K) being reached with n-heptane, inducing higher drive speeds of the expanded burnt gases. However, this effect is strongly attenuated with increasing equivalence ratios.
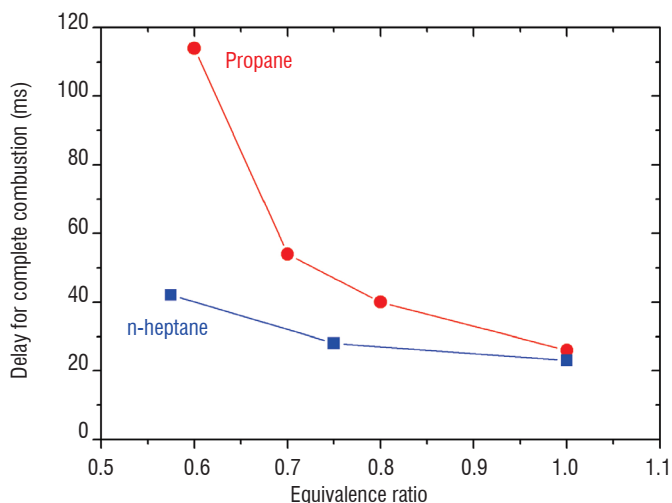

Figure 4 - Time needed to burn the entire mixture for various equivalence ratios (single point ignition).

P. Tardiveau et al [33] showed also that discharge filamentation had an effect on ignition. Ignition will be promoted by thinner plasma channels, where the energy density is higher. The constriction of filaments is more pronounced with n-heptane, but the gap distance is also a deciding parameter. The ability of the discharge to ignite air / HC mixtures, even at low equivalence ratios, is strongly correlated to the energy density that the discharge is able to release into the gas. When the experimental parameters are adjusted to make the discharge more filamentary, and then to increase the energy density, ignition is easier and combustion is faster [3, 33].

Lower concentrations of HC and shorter gap distances make the discharge more diffuse and the plasma channel radii larger, which reduces the energy density and does not promote ignition. Thus, it is necessary to increase the amount of electrical energy to make the combustion successful. Likewise, propane makes the discharge more diffuse. Apart from the specific kinetic properties of the two hydrocarbons, this should contribute to the fact that the ignition threshold energy is higher at a low equivalence ratio with propane than it is with n-heptane.

The diameter of the plasma channels is a key parameter that determines ignition. If it is too large, ignition becomes more difficult, or even impossible, whatever the energy released. In other words, it is more efficient for ignition to increase the energy density by varying the thinness of the plasma channels than by injecting more energy. This is an experimental finding deduced from a great number of fast imaging measurements. However, ignition always starts very close to the electrodes, at points where the electrical power density is higher.

The single nanosecond pulsed discharge is too short in time to heat the mixture by Joule effect. A nanosecond ultra-fast heating mechanism, involving the quenching reaction of nitrogen excited states on oxygen molecules, has been considered for other kinds of nanosecond discharge [2, 21]. However, spontaneous Raman spectroscopy measurements carried out on this discharge with 1.7% of propane (below flammability limit) and an energy of 30 mJ, tend to show that the temperature in the post-discharge at 1 mm from the tip remains under 500K [16]. Therefore, the reasons for ignition should be found partly in an efficient production of active radicals, such as $O(^3P)$ and OH.

Kinetic interpretation of the ignition of the air / n-heptane mixture

The effects of $O(^3P)$ radicals on the ignition of air / n-heptane mixtures were investigated with a simplified kinetic model of combustion [3]. Of course, other species are produced during the discharge, owing to electron collisions on molecules constituting the mixtures, in particular the first excited state of the oxygen atom, $O(^1D)$, various excited states of N_2 and O_2 (among which those that are metastable constitute an important energy reservoir), organic fragments (radicals and molecules) of the hydrocarbon, which could also come from the N_2 metastable quenching processes, etc. In our first approach, the goal is to highlight the role of the oxygen molecule dissociation yield producing $O(^3P)$ and to reveal specific kinetic routes for a low value of the initial temperature. Future work should be devoted to the role of species other than $O(^3P)$.

Together with specific reactions for air plasma [14] (but without electronically excited and ionized species), we used the low temperature reduced mechanism proposed by N. Peters et al [20], which includes the formation of ketohydroperoxydes following isomerization of the peroxy radicals. 37 species and 99 reactions were taken into account in all.
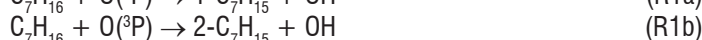
The model couples all balance equations for species densities to the temperature equation given by:

$$\frac{dT}{dt} = \frac{\sum_k^{N_{ü}} H_k \times [\dot{X}_k]}{\sum_k^{N_{ü}} C_{ü} \times [X]} \quad \text{with} \quad [\dot{X}_k] = \frac{d[X_k]}{dt} \quad (1)$$

where $[X_k]$ is the density, H_k is formation enthalpy and C_{Pk} is the heat capacity at constant pressure for each species k. The heat capacities and enthalpies are given by:

$$\frac{C_{Pk}}{R} = \sum_{n=1}^{5} a_{nk} T^{n-1} \quad \text{and} \quad \frac{H_k}{RT} = \sum_{n=1}^{6} \frac{a_{nk} T^{n-1}}{n} + \frac{a_{7k}}{T} \quad (2)$$

where the polynomial coefficients values a_{nk} were taken from H. Seiser et al [23]. This model enables the time evolution of the temperature and species to be calculated, under atmospheric pressure, for equivalent ratios Φ, ranging from 0.5 to 1.5 and initial temperatures ranging from 350 K to 750 K. Besides oxidation by molecular oxygen, we took into account initiation reactions between n-heptane and $O(^3P)$ or OH radicals:

$C_7H_{16} + O(^3P) \rightarrow 1\text{-}C_7H_{15} + OH$ (R1a)
$C_7H_{16} + O(^3P) \rightarrow 2\text{-}C_7H_{15} + OH$ (R1b)
$C_7H_{16} + OH \rightarrow 1\text{-}C_7H_{15} + H_2O$ (R2a)
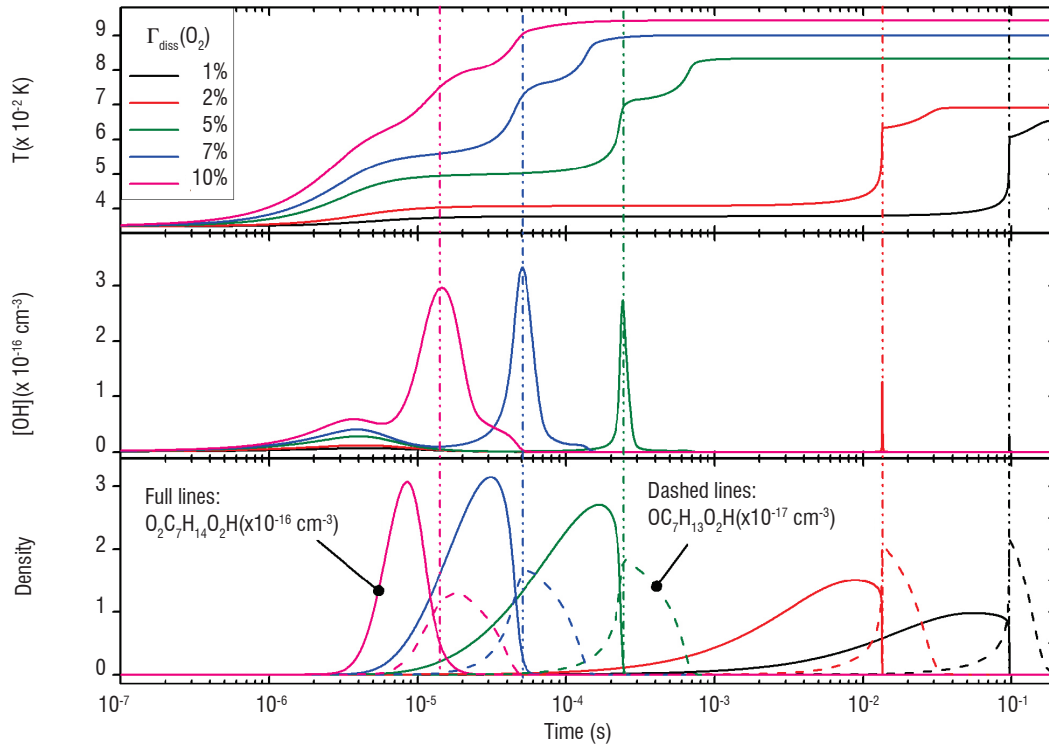$C_7H_{16} + OH \rightarrow 2\text{-}C_7H_{15} + H_2O$ (R2b)

Figure 5 - Calculated time evolutions of radicals (hydroxide OH and hydroperoxy heptyl peroxy $O_2C_7H_{14}O_2H$) and molecular ketohydrope-roxide ($OC_7H_{13}O_2H$) densities, and of temperature. Initial temperature: 350 K. O_2 dissociation yields $\Gamma_{diss}(O_2)$: given in the upper diagram.

The mechanism includes isomerization of peroxy radicals $C_7H_{15}O_2$ (resulting from the reaction of heptyl radicals 1- or 2-C_7H_{15} with O_2) forming hydroperoxy heptyl radicals, (R3), which leads to hydrope-roxy heptyl peroxy radicals by reaction with molecular oxygen, (R4):

$$C_7H_{15}O_2 \rightarrow C_7H_{14}O_2H \qquad (R3)$$
$$C_7H_{14}O_2H + O_2 \rightarrow O_2C_7H_{14}O_2H \qquad (R4)$$

By decomposition, this radical leads to the formation of an OH radi-cal and heptyl ketohydroperoxide $OC_7H_{13}O_2H$. This ketohydroperoxide acts as a degenerate branching agent yielding hydroxyl and ketohep-toxy radicals, (R5):

$$OC_7H_{13}O_2H \rightarrow OC_7H_{13}O + OH \qquad (R5)$$

The decomposition of ketoheptoxy radicals is also taken into account, (R6):

$$OC_7H_{13}O \rightarrow CH_2O + C_5H_{11} + CO \qquad (R6)$$

Molecular oxygen dissociation by electron collisions and quenching of the nitrogen excited states are an O-atom density source; this is why O_2 dissociation yields $\Gamma_{diss}(O_2)$ ranging from 0.1 % to 10 % were considered as input data. Figure 5 shows the results obtained with an initial temperature T_{ini} = 350 K and for a stoichiometric mixture ($\Phi = 1$).

Calculations were made for several O_2 dissociation yields, i.e., for seve-ral initial densities of O(^{3}P). It can be seen that this yield has a strong in-fluence, even though the maximum value chosen for our computations is rather moderate compared to some values published in the literature. Indeed, for a voltage pulse of 25 kV with a duration of 25 ns, operating at a repetition frequency of 10 Hz applied to the pin, the measurements performed by A. Lo et al [15] show that the O_2 dissociation reaches

almost 33% on the discharge axis, in agreement with the result of 50% given by G. Stancu et al [32] for a 10 kHz repetitive nanosecond pulsed discharge, in a pin-to-pin geometry. However, for the air / n-heptane mixture, the results plotted in Figure 5 emphasize that only 10% of the oxygen molecule dissociation leads to a strong reduction of the time required for a significant increase in the temperature.

The hydroxyl radical time evolution shows two peaks. One, between 3 and 4 μs, whatever the O_2 dissociation yield, increases with this dissociation rate. This first increase of OH, linked to the initiation pro-cess, comes from the balance between its generation by the reaction between n-heptane and the oxygen atom, (R1a) and (R1b), and its consumption, (R2a) and (R2b), which occurs mainly by reaction with n-heptane at this time.

The second peak in the OH time evolution is higher, and its time of occurrence is even shorter the higher the initial atomic oxygen density is. This increase is linked to the formation of heptyl ketohy-droperoxide $OC_7H_{13}O_2H$ resulting from the decomposition reaction of the hydroperoxy heptyl peroxy radicals $O_2C_7H_{14}O_2H$, which gene-rates one OH radical. Moreover, by reaction (R5), $OC_7H_{13}O_2H$ is also a source of OH.

In Figure 5, a rapid increase in the temperature is observed, along with the second OH peak and the ketoperoxide peak. This time, cor-responding to the maximum OH density, was chosen to determine the ignition time. It is identified by the vertical dotted-dashed lines in each diagram.

The computed ignition time as a function of the initial temperature for two equivalence ratios, 0.5 and 1, and two oxygen dissociation yields, 0.1 % and 5.0 % is shown in Figure 6. For a given initial tempe-rature, the time required for ignition to occur decreases when $\Gamma_{diss}(O_2)$ increases from 0.1 % to 5 %.

Also, for the two dissociation rates, the ignition time decreases when the initial temperature increases. Moreover, increasing the equivalent ratio from 0.5 to 1 entails a slight decrease in the ignition time. Note that for very short times, less than 2 μs, the two OH density peaks described in Figure 5 are not resolved in time. This prevents the determination of the ignition delay as defined above. In this case, this time is chosen corresponding to the maximum density of OH.
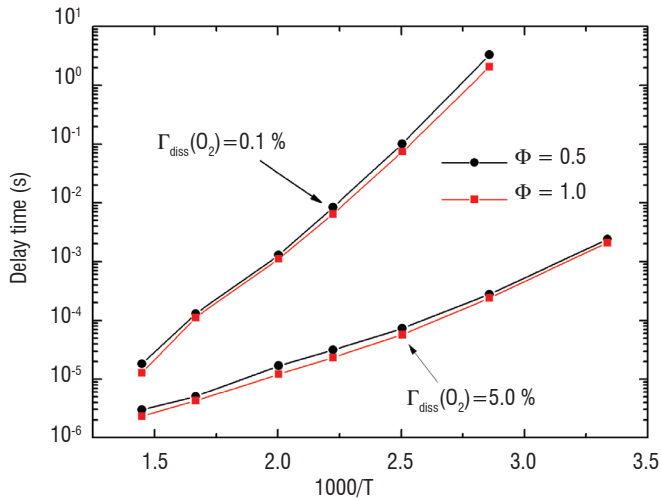

Figure 6 - Computed time required for the ignition of the air / n-heptane mixture as a function of the initial temperature (in K) for two values of the equivalence ratio Φ and of the dissociation percentage Γ_{diss} of the oxygen molecule.

Conclusion

The cold and non-equilibrium plasma generated by a single nanosecond pulsed overvoltage can be used to ignite various kinds of quiescent air / fuel mixtures. It shows several advantages, compared to classical spark gap devices: ignition of lean mixtures with low electrical energy consumption, extended flame kernels or several points of ignition and reduction of the ignition time.

A comparative study between propane and n-heptane emphasizes that less energy is required to ignite n-heptane and more specifically at low concentrations. The time required for the air / n-heptane mixture to burn completely is also reduced compared to propane for an equivalence ratio lower than one. Modeling shows that the oxygen atoms produced in the plasma appear to be a key species for these phenomena and, in the case of n-heptane, ketohydroperoxides, which are efficient combustion chain propagators through the generation of the hydroxyl radical, play a major role in the triggering of the combustion by the low temperature discharge.

Some of these results may be considered interesting for engineers working on the optimization of ignition devices, owing to the increasingly more important environmental constraints. As regards automobile engines, the implementation of a nanosecond corona discharge device on a spark plug has already been done some years ago [7] leading to the consideration of promising development.

As regards the interpretation of measurements, future work should be devoted to the role of species other than the oxygen atom in its ground state for the ignition of the air / n-heptane mixture. In particular, the production of the excited state $O(^1D)$ by the discharge deserves to be taken into account, because this state is much more reactive with hydrocarbons than $O(^3P)$. In addition, the development of a physical model describing the temporal evolution of the local reduced electric field E/N in the pin-to-plate gap, and consequently the spatial distribution of the energy deposition in the nanosecond corona discharge, should be of great interest if the reduced kinetic scheme proposed can be included in such a model. Such a physical model is particularly important, in order to correctly predict the local densities of the reactive species produced and thereafter the ignition of the combustible mixture at various locations between the electrodes ∎

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Acronyms

HC   (hydrocarbon)
HV   (high voltage)

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