Ignition delay and MIE measurement for n-decane/air mixture induced by laser-spark

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1 Introduction

Over the last decades the ignition of combustion using a laser spark has received a lot of attention [1-2] to replace the conventional electric spark plug. Recently this interest is also considered for combustion engines applications [3-4] due to the greater control over the timing and locations of ignition and its non intrusive nature [2]. In this field of activity parameters such as Minimum Ignition Energy (*MIE*) and ignition delay are key points to characterize the ignition of the studied reactive mixture. In propulsion applications fuel used are a mixture of various chemical species which can be problematic for fundamental and numerical studies [5]. As a consequence authors have focused their efforts on the n-decane/air mixture because n-decane is a part of the multi-components surrogates of kerosene. At authors knowledge there is a lack of data for this mixture ignited by a laser source at atmospheric pressure. The present work reports experimental measurements of MIE and ignition delay for n-decane/air mixture for different equivalence ratio with purity about *99.9 %* for the n-decane.

In this work, the measurement of the minimum ignition energy for n-decane/air mixtures are performed using a laser-induced spark ignition experimental system previously developed in [6] and upgraded in [7] for absorbed energy measurements. The next section is devoted to the presentation of the experimental apparatus and procedures used to determine the minimum ignition energy and ignition dealy of n-decane/air mixture. The third section is dedicated to the results and discussions. The last one is the conclusion.

2. Experimental device and procedures

In this work, a new step is achieved in the use of this apparatus with the contribution of a spectrometer to perform ignition delay measurements. A sketch of the apparatus is given in Figure *1*. Key aspects of the apparatus are reminded here. For a detailed presentation one can refers to [6-7].
The combustion chamber is a cylindrical vessel of $10^{-3}\ m^3$ ($L = 0.2\ m$ *and* $D = 0.8\ m$) filled with synthetic air (*80%* N_2, *20%* O_2, CO_2 *<1 ppm, CO ppm,* H_2O *< 3 ppm,* C_nH_m *< 0.1 ppm*) and liquid n-decane provided by *ALFA AESAR (A14732.AP as reference*) with a degree of purity of *99.9 %* following the partial pressure method.

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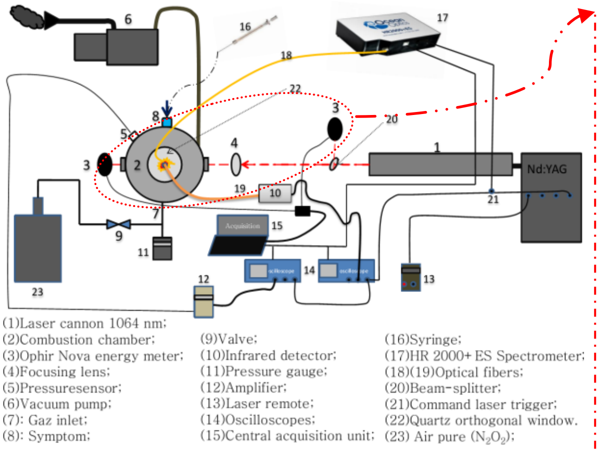

Figure *1*. Schematic diagram of the used apparatus.

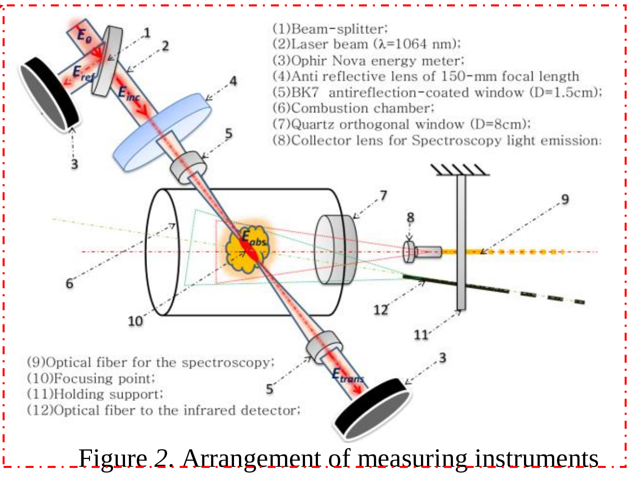

Figure *2*. Arrangement of measuring instruments and the principle of operation.

The reactive mixtures are ignited by the mean of a laser beam provided by a Q-switched Nd:YAG laser (Quantel Brilliant) operating at *1064* nm with pulse duration of *4.48* ns. The laser beam enters and exits the vessel through two antireflection coated windows.

The incident energy coming from the laser and transmitted through the mixture are measured by two Ophir Nova energy meters, see Figure *2*. The incident energy (E_{inc}) is calculated thanks to the first energy meter located behind the beam splitter, see Eq *1*:

$$E_{inc} = E_{ref}\left[\frac{(1-\rho)}{\rho}\right] \qquad (1)$$

With (ρ) the reflection coefficient of the beam splitter.

Then the energy absorbed by the plasma is calculated knowing the transmission coefficient (α) of the two antireflection coated windows and the measurement of the transmitted energy (E_{trans}), Eq *2*:

$$E_{abs} = \alpha E_{inc} - E_{trans} \qquad (2)$$

Both coefficients (ρ) and (α) depends on the atmospheric conditions. As a consequence they were calibrated every day and for every change of the energy of the laser source. Slight variations were observed for both ($\rho \approx 0.016 \pm 0.001$) and ($\alpha \approx 0.96 \pm 0.02$). During the calibration process, the reactor was under vacuum.

During experiments the ignition delay is performed with an infrared detector sensor (H250 SIEMENS SF) the collected signal is amplified before being recorded by a numeric oscilloscope. This sensor is used for two reasons: firstly it has a short response time of *10* ns which is largely consistent with the time scales of studied phenomena and secondly it has a maximum sensitivity in the near infrared $\lambda_{max} = 850$ nm with a sensitivity ranging from *400* nm to *1100* nm. This detector is used to detect the spark and the flame emission light from the laser for each trial of ignition. Also, at the same time we use an ocean optics HR*2000*+ES spectrometer to follow the induced spark and the flame light emissions with a wavelength resolution of *0.5* nm and an acquisition frequency of *500* Hz.

The study is carried out for *6* equivalence ratio (*0.65*, *0.9*, *1.1*, *1.3*, *1.6* and *2*) and *8* incident energies coming from the laser source (from *10* to *80* mJ with a *10* mJ step), for a total of *2400* shots.

3. Results and discussion

3.1 MIE and First estimation of Ignition delay

In order to compare our results to the data of Strozzi et al [5], MIE measurements are summed up in Figure *3*, using the approach developed in [8]. Before doing any comparisons, one can note that the obtained curve has the typical U-shape. In [5] authors worked with the same conditions of pressure and temperature and with a comparable range of incident energies and equivalence ratio, nevertheless the purity of n-decane was only *94 %*. It clearly appears that for lean mixtures the n-decane with the lowest degree of purity needs to absorb less energy to ignite while for rich mixtures it is the one with the highest degree which needs to absorb less energy. Moreover, as in [5], the studied mixture was not ignited for the leanest equivalence ratio. One can note that this equivalent ratio ($\Phi = 0.65$) is close to the Lower Flammability Limit ($\Phi = 0.55$) calculated using experimental correlation given in [9].

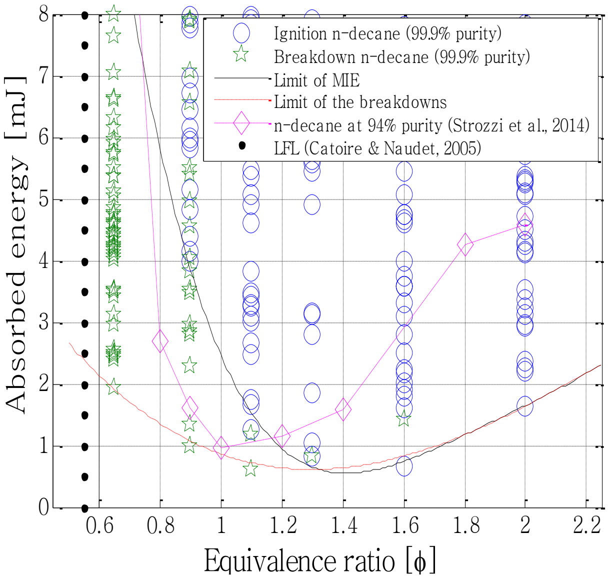

Figure *3*. MIE and minimum breakdown energy of n-decane/air mixture.

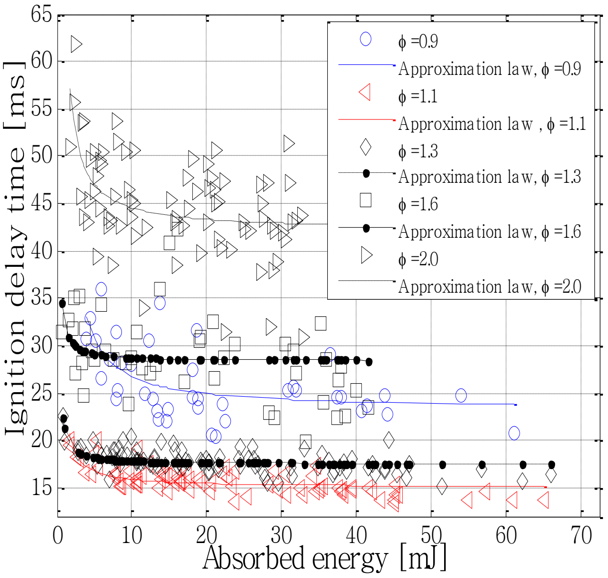

Figure.*4*. Ignition delay time as a function of absorbed energy for different ratios.

Ignition delay parameter is measured, in a first approach, with a fast infra-red detector and all the results are approximated using a based Arrhenius law for each equivalence ratio as a function of the absorbed energy (see figure.*4*). The dispersion of this parameter is a function of the equivalence ratio (Φ), we recorded a short ignition delay time and low dispersion near the stoichiometry ($\Phi = 1.1$; $13.43 < \tau$ (ms)< 20.12) and we have inverse amplification effect far from this operating point for Ex: ($\Phi = 2$; $30.9 < \tau$ (ms) < 61.78) (see the figure.*4*). this large dispersion is due to the different mode of ignition because we have one mode with big concentration of oxygen O_2 than probably we have a big chance to ionize molecules of oxygen than molecules of $C_{10}H_{22}$ and in this case we have production of OH^* and formaldehyde CH_2O for this regime, ignition needs more time to produce a sufficient amount of electrons avalanches and ions to start the combustion. The same thing for a high concentration of n-decane in high ratios where we have more probabilities to create radicals of CI^*, CII^* and CH^* those who undertake and maintain the chemical chain reactions. But at the stoichiometric state we have an equilibrated creation of different radicals fractions which provides an almost perfect balance to quickly begin and maintain the ignition.

The mathematical model which we devlop from the expérimental data to retrieve the ignition delay time as a function of the absorbed energy is given by Eq. *3*. And the two parameters *A* and *E* are given in Table.*1*.

$$\tau = A\exp\left[\frac{E}{E_{abs}}\right] \qquad (3)$$

Table.1. Experimental values of (*A*) and (*E*).

ϕ	*A*	*E*
0.9	*23.24*	*1.40*
1.1	*15.04*	*0.54*
1.3	*17.49*	*0.20*
1.6	*28.36*	*0.13*
2.0	*42.16*	*0.55*

Where we recall that all the values of *A* and *E* are calculated at atmospheric pressure and initial temperature of *347 K.*

3.2 Estimation of Chemical induction delay and spectroscopic study

This chemical parameter measures the delay time from which we have appearance of a radical radiation or a chemical species related to a fixed wavelength. For this study it is judicious to have a spectrometer or a photo- multiplier device with monochromators filters. In our case we use the HR *2000*+ES provided by Ocean Optics which enable the acquisition of one spectra in the range *200* to *1100* nm every *1* ms.

As showing in Figure.*5*, the chemical delay is estimated by the exploitation of the spectroscopic measurements which represents one case of laser spark ignition and the flame emission spectroscopy as function of time. The integration time of one spectrum is about *2* ms.

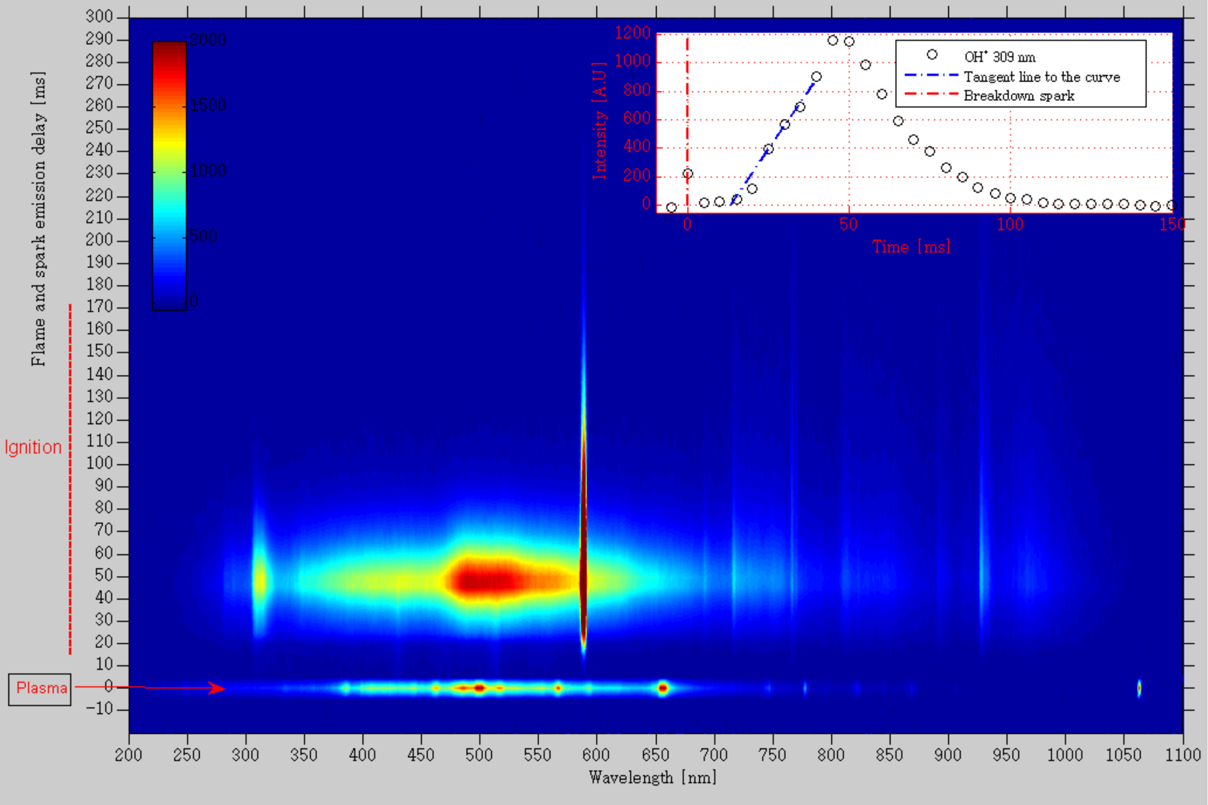

Figure.*5*. Laser spark ignition and flame emission delay time using spectroscopy ($\phi=1.1$; $E_{inc}=60mJ$).

At the check-looking full spectrum, we can see the emitted bands during the breakdown from *0* ms to about *2.5* ms in Figure.*5*. The laser beam light emission is clearly visible at *1064* nm and give the start of the process. At *778.2* nm we detect an oxygen atom ionized once (*OI*),
The ionized hydrogen phase in its alpha state is largely visible at *657* nm with the others states of the ionized nitrogen, oxygen and carbon (*NI, NII OII and OIII CI*) it is thanks to them that creation of radicals OH^*, CH^* *and* CII^* is guaranteed. Their traces are distinguished beings just after breakdown by low intensity line emission.

These measurements enable us to perform some analyses on the different chemical delay times represented by a fixed emission wavelengths of four radicals CII^*, OH^*, CH^*, CI^*. The procedure is to follow a specific wavelength in time, taking its tangent between *20 % and 80 %* of the maximum signal strength and make it cut the time-axis. The breakdown is indicated by the pick of the *1064* nm laser beam. Finally the estimated chemical delay will be the difference between the time recorded by the intersection of the tangent line with the time-axis and the first apparition of the focused laser beam. Variation of the corresponding ignition delay according to equivalence ratio are plotted in Figure.*6*.

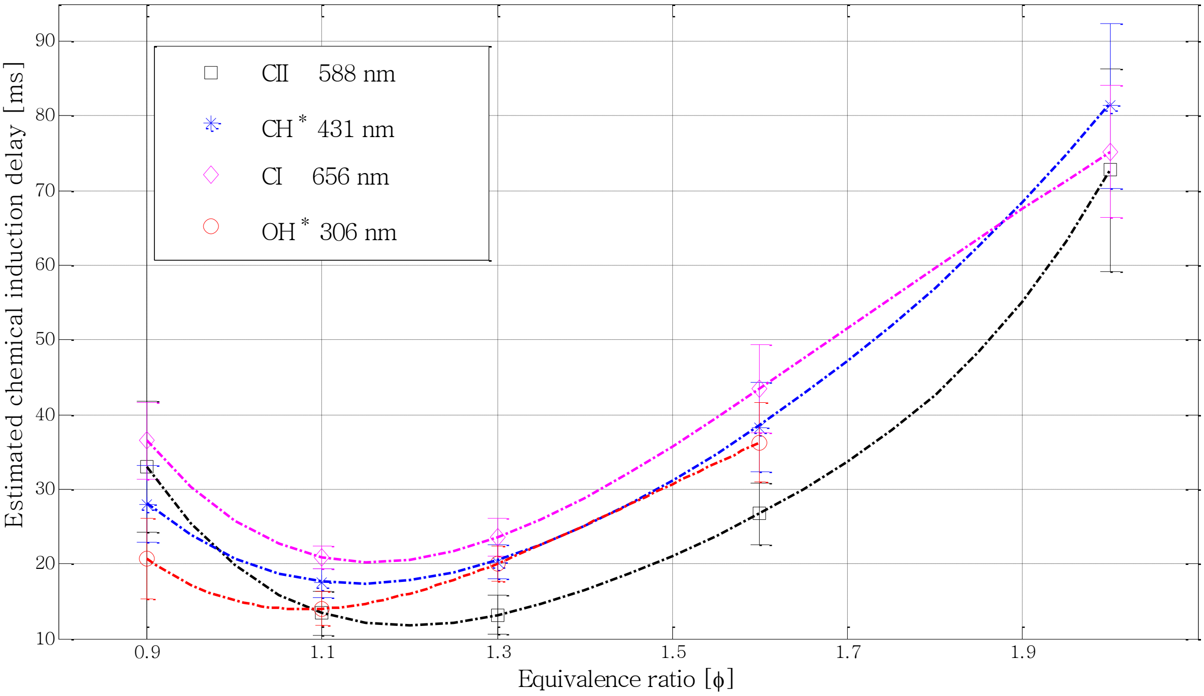

Figure.*6*. Estimated chemical induction delay as a function of equivalence ratio.

Comparison between estimated chemicals delays times shows that for the lean mixtures, radical OH^* is the first to make apparition next it follows by the radical CH^* than CII^* and CI^*. Near the stoichiometry the radical OH^* and CII^* have the same time of apparition and followed by the radical CH^*. For higher equivalence ratio, CII^* is ahead other radicals, it worth noticing that the OH^* and CH^* radicals appears at about the same time. The CI^* specie is the last to appear at $\phi =2$ where it anticipates the radical CH^*. The lower chemical induction delay is recorded for CII^*, near stoichmetriy and is about *11.74* ms with a minor error dispersion than for the OH^* at *14.01* ms followed by the CH^* at *17.33* ms and finally by CI^* with *20.21* ms. Far away from the stoichiometry, the dispersion error intensifies with the increasing and the decreasing of equivalence ratio. The observations from these results can be a significant help for developing correct kinetic schemes. It should also improve numerical simulations to have the actual induction time in the same conditions of experience (Laser ignition).

4 Conclusion

This experimental study allowed the measuring of the minimum ignition energy (*MIE*) and the Ignition delay time (*IDT*) in a cylindrical chamber at fixed atmospheric pressure and an initial temperature of *347K* for six chosen equivalence ratios.

In the first part, minimum ignition energy is determined for pure n decane, the MIE have been compared with the data of Strozzi [5]. This comparison allows us making in evidence the influence of impurities in the laser induced ignition. As shown previously impurities act as a catalyst for the ignition at lower ratios and as inhibitor for higher ratios. From this observation, it is noticeable that a small amount of impurities about *6 %* can play a significant role on the characteristics of ignition.

Then we have focused the study on the measuring of the ignition delay time with a fast infra-red detector and we have proposed a based Arrhenius law which give the variation of this parameter as function of the equivalence ratio and absorbed energy. From this section we have found that the dispersion value of the measuring of ignition delay time increases at lower and higher equivalence ratios and is minimum near the (ϕ=*1.1-1.2*) range. We have also demonstrated the effect of the absorbed energy on the ignition delay time. For low absorbed energies the ignition time tends to infinity and for considerable energy absorbed the ignition time tend to a limit which is a function of the stoichiometry.

In the same case the different chemical induction delay time have been monitored for 4 radicals (CII^*, OH^*, CH^*, CI^*) by spectroscopic measurements. This part provides a validation of the first results achieved within the IR detector because we recorded the same influence of the equivalent ratios. The estimated chemical induction delay of different species can be shorter as it can be longer given like a function of the mixture ratio.

This study belong a global researches which also plan to determine the influence of addition of argon and moisture on ignition condition [10]. In a next future, we plan looking at the influence of initial pressure, temperature condition and the use of shorter focal lengths and we will try to make a global law which relay these parameters to the physical and thermodynamics proprieties of the mixture Air/fuel(n-decane).

Acknowledgements

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References

[1] Klimkin VF, Soloukhin RI, Wolansky P. (1973). Initial stages of a spherical detonation directly initiated by a laser spark. Combustion and flame. 21:111.

[2] Phuoc TX. (2006). Laser-induced spark ignition fundamental and applications, Opt.Lasers Eng. 44:351.

[3] Morsy MH. (2012). Review and recent developments of laser ignition for internal combustion engines applications. Renewable Sustainable Energy Rev. 16:4849.

[4] Griffiths J, Riley M, Kirk A, Borman A, Lawrence J, Dowding C. (2014). Lean burn limit and time to light characteristics of laser ignition in gas turbines. Optics and Lasers in Engineering. 55: 262.

[5] Strozzi C, Gillard P, Minard JP. (2014). Laser-induced spark ignition of gaseous and quiescent n-decane–Air Mixtures. Combustion Science and Technology. 186:1562.

[6] Tihay V, Gillard P, Blanc D. (2012). Ignition study of acetone/air mixtures by using laser induced spark. Journal of Hazardous Materials. 209-210:372.

[7] Rudz S, Chetehouna K, Strozzi C, Gillard P. (2014). Minimum Ignition Energy Measurements for α-Pinene/Air Mixtures. Combustion Science and Technology. 186:1597.

[8] Moorhouse J, William A, Maddison TE. (1974). An investigation of the minimum ignition energies of some C1 to C7 hydrocarbons. Combustion and Flame. 23:203.

[9] Catoire L, Naudet V. (2005). Estimation of temperature-dependent lower flammability limit of pure organic compounds in air at atmospheric pressure. Process Saf. Prog. 24:130.

[10] Gillard P, Mokrani N, Rudz S, Strozzi C, Tihay V. (2014). Effect of moisture and argon on laser ignition of ethanol and decane, ; (X ISHPMIE) Tenth International Symposium on Hazards, Prevention, and Mitigation of Industrial Explosions, Bergen, 12-14 June 2014.