Laser Induced Spark Ignition of Gaseous and Quiescent n-decane - air Mixtures
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1 Introduction
Ignition of jet fuels has received much attention over the last decades. If the performances of aeronautical engines represent a crucial issue, numerous research efforts concern the risk of explosion as well. In particular, civil aviation is concerned as several works have been performed to improve safety, for instance following the TWA 800 accident. Efforts have also been related to military applications like vulnerability or fire safety studies. Some of these studies aim at characterizing properties like volatility, flashpoint, composition and ignitability of different fuels like Jet-A, JP8, other kerosenes or related surrogates [1,2,3]. In this respect, n-dodecane and in a lesser extent n-decane have been considered as single component or parts of multi-components surrogates of kerosenes [4,5]. Determining their ignition properties is therefore of interest for the different fields of application cited above.

If the ignition characteristics are generally determined by using electrical spark devices [6], laser spark ignition has been widely used for this purpose as well [7]. It is also the case for fuels representative of aeronautic applications, see for instance Lee et al. [1] who determined Minimum Ignition Energies (MIE) of propane, dodecane, Jet-A fuels via laser spark ignition. Interest in this ignition technique has also increased in recent years because of its many potential advantages over conventional ignition systems. The major benefits [7] are a greater control over the timing and locations of ignition and their non-intrusive nature. Moreover, these features are interesting for flame holding in rapid reactive flows. Therefore laser ignition is under consideration as a potential candidate for future propulsion applications.

The present manuscript reports an experimental study of the behaviour of gaseous n-decane – air mixtures submitted to laser sparks. In particular the breakdown and ignition probabilities are determined for different equivalence ratios. The MIE is determined following the approach of Moorhouse [8], it is compared to the literature.

2 Experimental devices and procedures
The laser beam is provided at 1064 nm by a Q-switched Nd:YAG laser (Quantel Brilliant). The Gaussian beam (quality factor $M^2 = 1.95$, diameter $d = 6$ mm) is focused in the center of the...
combustion chamber by a 150 mm focal length lens, see Fig. 1. As the laser features a pulse duration of 4.48 ns, a high irradiance is provided. As a result, multi-photon ionization and the subsequent electronic avalanche occur. These processes are at the origin of the non-resonant laser breakdown phenomenon.

The incident energy \( E_{\text{inc}} \) and the energy transmitted \( E_{\text{tr}} \) through the mixture are monitored by two Ophir Nova energy meters (2) and (3) coupled with 10-AP thermal sensors. The subtraction of these two values leads to the energy absorbed by the plasma \( E_{\text{abs}} \). It is worth noticing the values reported in the present study are corrected to account for the windows absorption. They correspond to the phenomena occurring into the gaseous mixture contained within the chamber. A photodiode detector is used to monitor the laser spark emission.

![Figure 1. Sketch of the experimental apparatus](image)

The combustion chamber is cylindrical \((l = 200 \text{ mm}, D = 80 \text{ mm})\) with a volume of 1 L. It is fitted with two quartz windows in order to visualize the laser spark and the combustion process. They are orthogonal to two anti-reflection coated BK7 windows \((d = 15 \text{ mm})\) allowing the passage of the laser beam. The reactive mixture is obtained from real air and from liquid n-decane with a degree of purity of 94%. The mixture preparation follows the partial pressures method. The chamber is first placed under vacuum. Then the liquid fuel is directly injected into the combustion chamber through a septum using a micrometric syringe. Air is then admitted with a flow rate of approximately 2 L/min. The equivalence ratio is calculated from the volume of liquid n-decane and the total pressure \( P_0 = 1 \text{ bar} \). The combustion chamber and the gas feeding pipe are heated at \( T = 347 \text{ K} \) to allow the fuel vaporization over a wide range of fuel equivalence ratio. The chamber is filled with air and placed under vacuum twice between consecutive reactive experiments in order eliminate residual products of combustion and residual moisture. The walls and windows of the chamber are periodically cleaned. A Cormak 9555 differential manometer with one mbar accuracy is used for the static pressure measurements. During combustion experiments, the pressure evolution is monitored by a piezoelectric transducer (Kistler 603B) with a charge amplifier (Kistler 5011).

### 3 Results and discussions

The study first reports the results related to an inert decane-nitrogen mixture. In a second part, the laser breakdown and ignition of a n-decane - air mixture are considered in terms of incident energy. Finally the focus is on absorbed energies and in particular on the Minimum Ignition Energies (MIE).
The non resonant laser breakdown phenomenon is first studied by considering an inert mixture of n-decane and nitrogen. The partial pressures are the following: $P_{nC_{10}H_{22}} = 974$ mbar and $P_{C_{10}H_{22}} = 26$ mbar. The latter is equal to that of a stoichiometric n-decane - air mixture. It enables the study of the laser breakdown independently from the combustion process.

![Image of laser breakdown data obtained with a n-C$_{10}$H$_{22}$ – N$_2$ mixture ($P_0 = 1$ bar, $P_{C_{10}H_{22}} = 26$ mbar, $T_0 = 347$ K). Left: Transmitted energy versus incident energy. Right: Mean absorbed energy versus incident energy.]

Figure 2 depicts the incident energy $E_{inc}$ versus the transmitted energy $E_{tr}$. Depending on the incident energy, different behaviors are observed: in a first zone, no breakdown occurs and therefore $E_{tr} = E_{inc}$. A transition zone is then observed for intermediate energy levels ($E_{inc} \approx 32$ to $52$ mJ) where the laser shots do not systematically lead to laser breakdown. For higher incident energies, all the shots are followed by a laser spark. In the last two cases, the absorbed energy fluctuates for a same value of the incident energy. Such fluctuations result from the stochastic nature of the non-resonant breakdown phenomenon itself. They can also be caused by the presence of aerosols, characterized by a lower ionization potential than gases. In that case, larger fluctuations are observed for long focal lengths [9].

In conclusion, the results shown in Fig. 2 underline the need for statistical characterizations of the breakdown and the subsequent ignition when considering laser ignition for practical applications.

The mean absorbed energy is also reported in Fig 2. The values tend to stabilize at high incident energies, revealing the plasma reaches a saturation regime. It corresponds to an increase of the size of the plasma whereas the temperature and density are stabilized. This phenomenon was previously reported for instance in [9,10]. The saturation occurs about three times the energy corresponding to the end of the transition zone (52 mJ), an observation in agreement with the data reported in several studies for different focal lengths and gaseous mixtures [9,10].

**Ignition and breakdown of a n-decane – air mixture**

In this section, ignition statistics are under consideration for reactive mixtures of n-decane - air. Breakdown and ignition probabilities are calculated by performing 20 reactive experiments for each incident energy.

Figure 3 displays the ignition probability functions in function to the incident energy $E_{inc}$ for different equivalence ratios $\Phi$. From a qualitative point of view, they present similarities with functions obtained from electrical spark ignition. The stoichiometric mixture displays the highest ignitability, whereas higher incident energies are necessary to ignite lean or rich mixtures. For the leanest mixture ($\Phi = 0.65$) the probability values are shifted towards particularly high energies, as the equivalence ratio is close to the lower flammability limit ($\Phi = 0.6$). The shift is lower for rich mixtures as the equivalence ratio remains far from the upper flammability limit ($\Phi = 4.3$).
Figure 3: Ignition probabilities for lean (left) and rich (right) n-decane – air mixtures.

Figure 4: Evolution of $E_{50}$ based on incident energy for different n-decane – air mixtures.

Figure 5: Ignition and breakdown probabilities for different n-decane – air mixtures.

Figure 4 illustrates as well these trends. It depicts the evolution of the incident energy leading to 50% of successful ignitions with respect to fuel equivalence ratio: $E_{50} = 72.6$ mJ, 30.7 and 39.2 mJ are respectively obtained for the values $\Phi = 0.65, 1$ and 2. These values result from linear interpolation.

Furthermore, Fig. 3 shows that the probability function rises sharply at intermediate and high probabilities, whereas the slope is lower at low probabilities. Such a plateau at low probabilities was observed in previous studies [10,11].

Figure 5 reports both breakdown and ignition probabilities for the n-decane - air mixture. For lean mixtures, the ignition probability (solid lines) is equal or lower than the breakdown probability (dotted lines). This effect is particularly pronounced at $\Phi = 0.65$, a value close to the lower flammability limit ($\Phi = 0.6$). Morsy and Chung [12] evidenced complex flow patterns with shock and rarefaction waves that are at the origin of the formation of contrarotating toroidal rings. Following Phuoc et al [7], the subsequent high rate of stretch may inhibit the development of the flame kernel for very lean mixtures. These interpretations are consistent with the large gap observed in Fig. 5 between the breakdown and ignition probabilities. This effect might be enhanced by the fact n-decane should promote the plasma formation in comparison to air. Indeed, the ionization potential of n-decane equals 9.73 eV, which is lower than that of air (15.7 eV).

For higher equivalence ratios, each breakdown leads to ignition and hence the same probabilities are obtained for both phenomena. Such behavior was already observed in a previous study [10]. This also means the plateau observed at low ignition probabilities seems to be rather the result of a breakdown related phenomenon than to be caused by a process that takes place during the flame kernel development.

Figure 6 defines the domains of systematic misfire/ignition in terms of incident energy and equivalence ratios. This overall representation summarizes the results in a relevant way for an application to practical combustion systems. Depending on the values of equivalence ratio, it clearly appears more than 50 to 90 mJ must be delivered by the laser (incident energy) to perform a systematic ignition of a quiescent mixture in the conditions of the present study.
From a more fundamental point of view, it is appropriate to characterize the reactive mixture in terms of absorbed energy, as significant fluctuations occur for constant incident energies. It is particularly true for Minimum Ignition Energies (MIE) estimation.

The results of the set of experiments are summarized in Fig. 7 following the approach of Moorhouse [8]. The MIE corresponds to the minimum energy leading to a successful ignition experiment. Low values (0.95 - 1.6 mJ) are obtained for equivalence ratios included in the range $\Phi = 0.9 - 1.4$. A sharp increase is observed on the lean side as the mixture composition is close the flammability limit. The increase is moderate on the rich side. The U-shaped curve is similar to that obtained for different mixtures [7]. The MIE value equals 0.95 mJ, which is in the usual range for alcanes – air mixtures when using electrical spark as an ignition process (isooctane 1.35 mJ, heptane 0.24 mJ, methane 0.28 mJ, see [13]). The values are also consistent with the fact MIE of hydrocarbon-air mixtures obtained by non-resonnant laser spark are in general slightly higher than that obtained by electrical spark near the stoichiometry, a closer agreement being obtained for lean and rich mixtures [7].

Furthermore, Dietrich et al. [14] studied the spark ignition of bidisperse n-decane spray. An ignition energy equal to 0.5 mJ is reported at $\Phi = 1.5$. This value is lower than that of the present study, which is expected as it is obtained for a higher oxygen molar fraction (37%).

Lee et al [1] performed laser ignition experiments with a dodecane - air mixture for different temperatures at the fuel vapor saturation. A MIE equal to ~1 mJ is reported at atmospheric pressure and $T_0 = 330$ K. This MIE value agrees with the present measurements. Nevertheless Lee et al obtained this minimum energy at $\Phi = 3.5$ while the minimum measured for n-decane in the present study corresponds to stoichiometry. This difference is surprising as the equivalence ratio corresponding to the MIE is generally slightly shifted from the stoichiometry to the rich side for an increasing fuel carbon number. Here the difference is significant while the carbon numbers are relatively close together. Ignition energies are also compared at constant equivalence ratio $\Phi = 2$: a value close to 9.5 mJ is reported by Lee et al. [1] ($T_0 = 310$ K), while in the present study the value 4.4 mJ is obtained at $T_0 = 347$ K. Despite the different initial temperatures, the orders of magnitude of ignition energies remain in agreement for the same fuel equivalence ratio.
4 Conclusion

The present study provides experimental data related to gaseous n-decane – air mixtures submitted to non resonant laser sparks. In particular the breakdown and ignition probabilities are determined for different equivalence ratios. The domain of systematic ignition is determined in terms of energy delivered by the laser (incident energy) and equivalence ratio. A value of 0.95 mJ is determined for the Minimum Ignition Energy (MIE) following the approach of Moorhouse [8]. This value is consistent with the literature. The influence of other parameters like the initial pressure and the air moisture are currently under study and will be reported in the future.

To the author’s knowledge, previous works with n-decane - air mixtures essentially focus on spray ignition by electrical spark. Therefore the present work provides a relevant complement to the existing data. These results may be useful in the framework of hazard prevention studies but also for propulsion applications.

References


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