Journal of Hazardous Materials 209–210 (2012) 372–378

Contents lists available at SciVerse ScienceDirect

Journal of Hazardous Materials

journal homepage: www.elsevier.com/locate/jhazmat

Ignition study of acetone/air mixtures by using laser-induced spark

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ARTICLE INFO

Article history:
Received 24 November 2011
Received in revised form 12 January 2012
Accepted 12 January 2012
Available online 21 January 2012

Keywords:
Acetone
Laser breakdown
Ignition probability
Minimum ignition energy
Laminar burning velocity

ABSTRACT

The breakdown and the laser-induced spark ignition of acetone–air mixtures were experimentally studied using a nanosecond pulse at 1064 nm from a Q-switched Nd:YAG laser. The breakdown was first characterized for different mixtures with acetone and air. This part of the work highlighted the wide variation in the energy absorbed by the plasma during a breakdown. We also demonstrated that the presence of acetone in air tends to reduce the energy required to obtain a breakdown. Next, the ignition of acetone–air mixtures in the equivalence ratio range 0.9–2.4 was investigated. The probabilities of ignition were calculated in function to the laser energy. However, according to the variability of energy absorption by the plasma, we preferred to present the result according to the energy absorbed by the plasma. The minimum ignition energies were also provided. The minimum ignition energy was obtained for an equivalence ratio of 1.6 and an absorbed energy of 1.15 mJ. Finally the characteristics of the plasma (absorption coefficient and kernel temperature) were calculated for the experiments corresponding to minimum ignition energies.

1. Introduction

Acetone ($(CH_3)_2CO$) is an oxygenated hydrocarbon belonging to the ketone family. This colorless, volatile, flammable liquid is widely used in industry. It is mainly used as an industrial solvent, where it provides the active ingredient in many cleaning products, in the nail polish removers, in thinners of paint, glue or resin and in various degreasers. It is also used as a precursor to other chemicals. Acetone is indeed used in the manufacture of methyl methacrylate, employed in plastics and PVC industry and in the manufacture of bisphenol A (BPA) which is a raw material used in the production of epoxy and polycarbonate resins. The most common hazard associated with acetone is its extreme flammability. The acetone vapors are heavier than air and may travel a considerable distance. At temperatures above the flash point of acetone (−20 °C), air mixtures of between 2.5% and 12.8% acetone by volume can be ignited by source ignition such as flame or electrostatic discharges. To prevent the risk of explosion during industrial processes, knowledge of properties of inflammation such as minimum ignition energy (MIE) is required. Most studies on MIE use a capacitive spark discharge [1–7]. However, the MIE values obtained with this kind of apparatus are influenced by test conditions such as the properties of the electrical circuit. Given this, it seemed interesting to use another ignition source to perform the experiments for determining the MIE.

Interest in laser ignition has increased in recent years because of its many potential advantages over conventional ignition systems. The major benefits are a greater control over the timing and locations of ignition and their non-intrusive nature. There are generally four mechanisms by which laser radiation can ignite a combustible solid, liquid or gaseous mixture [8]: laser-induced thermal ignition, laser-induced photochemical ignition, laser-induced resonant breakdown ignition and laser-induced spark ignition. In the latter case, a laser beam with a sufficient irradiance (in order of 10^{10} W/cm^2) is sufficient to generate a spark plasma at the end of the laser pulse. This process begins with multi-photon ionization of gas molecules. This releases electrons, which absorb more photons via the inverse Bremsstrahlung process, increase their kinetic energy. The electrons liberated by this means collide with other molecules and ionize them. It leads to an electron avalanche and to the gas breakdown. A spark plasma of high temperature and high pressure is created. This extreme condition relative to the ambient gas leads to the development of a rapidly expanding shock wave that is of sufficient strength to ignite flammable mixtures [9].

The ionization of gases by focused laser beams has received considerable attention in order to understand the mechanisms leading to a breakdown. Several gases have been studied, such as air [10–16], rare gases [17–20], hydrogen [21–23] or methane [12–14]. The laser-induced ignition has also been investigated. Studies have

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doi:10.1016/j.jhazmat.2012.01.040

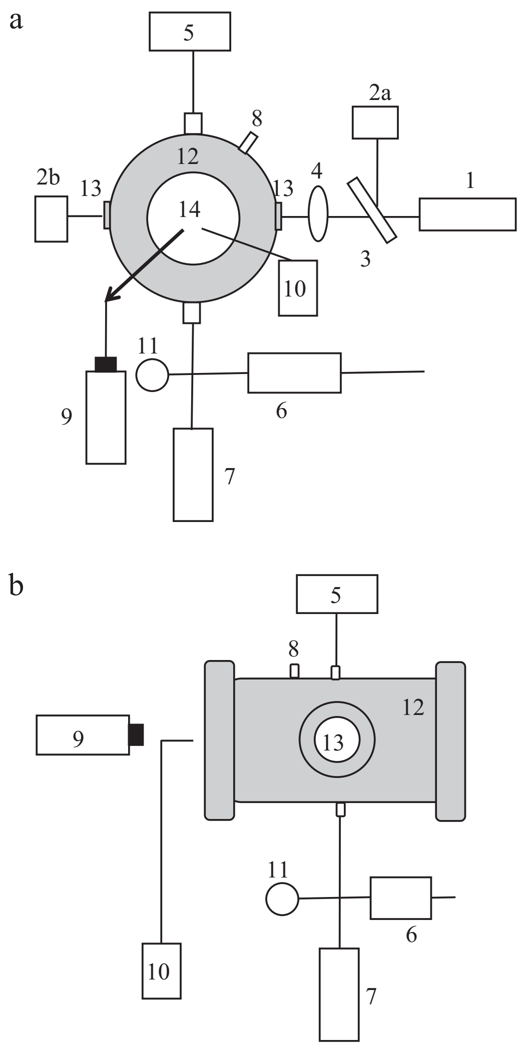

Fig. 1. Sketch of the experimental apparatus: (a) side view and (b) front view. (1) Nd:YAG laser; (2) energy meter; (3) beam splitter; (4) 150 mm focal length lens; (5) pressure transducer; (6) vacuum pomp; (7) gas inlet; (8) septum; (9) camera; (10) photodiode detector; (11) manometer; (12) chamber; (13) antireflection-coated window and (14) quartz window.

mainly focused on mixtures containing hydrogen [21–23], methane [24–28] or propane [29] although some studies have been also performed on hydrocarbon fuels such as dodecane, isooctane or Jet-A [29–31]. In these studies, the ignition characteristics are usually expressed according to the energy delivered by the laser. To our knowledge, few studies have examined the breakdown of acetone [32] and none has been conducted on laser-induced ignition for mixtures of acetone–air. Thus, to better understand the ignition of acetone–air mixture, this study investigates first the processes leading to the gas breakdown. Then, the energy required to ignite the mixture was determined as a function to the equivalence ratio. Finally, the characteristics of the plasma were calculated for the MIE.

2. Materials and methods

2.1. Experimental device

The experimental setup is shown in Fig. 1. A Q-switched Nd:YAG laser (Quantel Brilliant) operating at 1064 nm with pulse duration of 4.48 ns was used as the ignition source. A 150-mm focal length lens was used to focus the initial 6-mm-diameter beam. By assuming that the focal region of the beam is cylindrical, the spark size, in terms of radius r and length l, is given by [26]:

$$r = \left(\frac{2\lambda}{\pi}\right)\left(\frac{f}{D_0}\right) \quad (1)$$

$$l = (\sqrt{2}-1)\frac{\theta}{D_0}f^2 \quad (2)$$

Here f is the lens focal length (equal to 150 mm), D_0 is beam diameter (equal to 6 mm) and θ represents the beam divergence (equal to 0.44 mrad). For this study, the focal point characteristics are $r = 16.93\ \mu m$, $l = 683\ \mu m$ and the volume is equal to $\nu = 6.15 \times 10^{-13}\ m^3$.

The laser energy was controlled with a beam attenuator. Two energy meters (Ophir meter (NOVA) with a thermal sensor 10 A-P) were used to measure the beam energy before (2a) and after the chamber (2b). The laser energy before the chamber was obtained by using a beam-splitter that reflected 5% of the incident energy while transmitting the rest. A correction was performed on the data collected by the energy meters to account for the optical losses (absorption and scattering) of the experimental device.

The combustion chamber was cylindrical (8 cm diameter × 20 cm long) with a volume of 1 L. Two antireflection-coated windows in BK7 (15 mm diameter) allow the passage of the laser beam. Two orthogonal windows in quartz (80 mm diameter) were used to view the entire volume of the chamber. The visualization was performed with a camera recording 1000 frames per second. A photodiode detector was also employed to detect the spark emission light from the laser. A hole below the chamber was used for gas inlet and outlet and was equipped with a differential manometer (Comark C9555) for pressure control with an 1 mbar accuracy. Another hole above the chamber with a septum was employed for liquid injection. A pressure transducer (Kistler, Model 603B) combined with a charge amplifier (Kistler, Type 5011) was placed above the center of chamber to measure the dynamic pressure during the combustion.

This study investigated different gas compositions including air and acetone (Table 1) at ambient temperature (22 °C): pure acetone at 220 mbar (pressure below the saturation vapor pressure equal to 269 mbar at 22 °C), air at 1 bar and acetone/air mixtures at 1 bar with equivalence ratios between 0.9 and 2.4 to remain in the flammability range of acetone. Acetone (VWR) used in the experiment is commercially available and has a minimum purity of 99.5%. Before each experiment, the chamber was initially evacuated to remove residual gases and moisture. Acetone was then injected into the chamber using a syringe through the septum. The quantity was determined using the phase-equilibrium calculations (Clausius–Clapeyron equation). The saturation vapor pressure at 20 °C and the enthalpy of vaporization were taken equal to 247.4 mbar and to 31.3 kJ/mol, respectively. The vaporization of acetone was monitored with the differential manometer. After about 5 min, the partial pressure of acetone was equal to the expected value (Table 1) reflecting the fact that the acetone was completely vaporized. Air was then introduced into the chamber with a flow rate of about 20 L/min until the pressure reaches 1 bar. We again waited 5 min before starting the tests to ensure that the turbulence had disappeared and that the concentration of acetone/air mixture was uniform in the chamber.

2.2. Probabilities and thresholds

The probability of breakdown or ignition was defined as the ratio of positive trials and the number of shots. For breakdown, a positive test corresponded to the formation of plasma, which was characterized by a flash of light and a sharp acoustic sound. For ignition, a positive test involved the formation of a flame front, an increase

Table 1
Pressures of acetone and air used in the study.

		Air	Equivalence ratio for Acetone/Air mixtures									Acetone
			0.9	1	1.2	1.4	1.6	1.8	2	2.2	2.4	
Pressure (mbar)	Air	1000	955	950	941	932	923	914	905	896	888	0
	Acetone	0	45	50	59	68	77	86	95	104	112	220

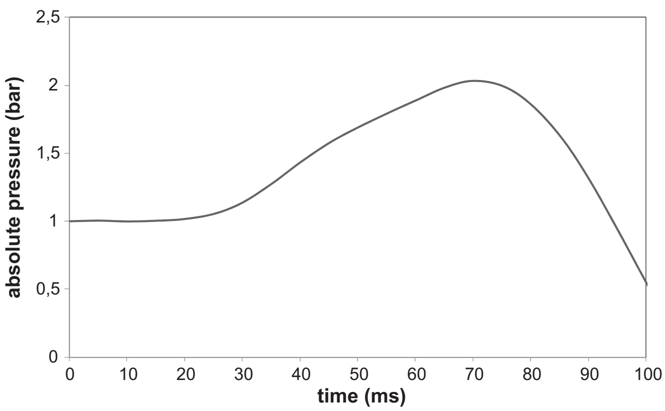

Fig. 2. Typical pressure profiles of an ignition event.

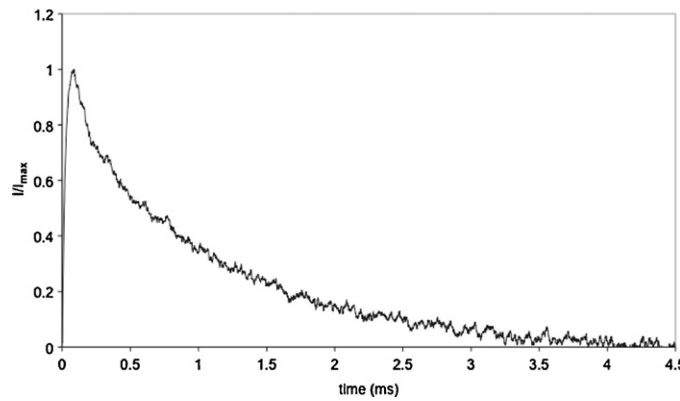

Fig. 4. Light intensity during a breakdown in acetone.

of pressure inside the chamber (Fig. 2) and the combustion of the mixture. Four thresholds were used to characterize the results:

- The energy E_{100} representing a systematic breakdown/ignition that is to say with a probability equal to 100%.
- The energy E_{50}. At this threshold, one-half of the incident laser pulses causes the breakdown/ignition.
- The energy E_1 corresponding to a breakdown/ignition with a probability of 1%.
- The minimum ignition energy (MIE) defined by using the approach of Moorhouse et al. [3]. It consists to identify a borderline between ignition and no-ignition by varying the amount of energy deposited in the mixture, for several equivalence ratios.

3. Results and discussion

3.1. Plasma description

Fig. 3 shows a typical image of breakdown. The laser beam comes from the right side. The plasma has an ellipsoidal shape, which is consistent with previous studies on the breakdown [11,15,24]. This shape is due to spherical aberrations in the lens, which produce plasma with multiple points. The images show also that the initial plasma expands opposite to the laser's incident direction. The color of the breakdown varies depending on the gas composition. For air, the laser-induced spark is very bright and is blue.

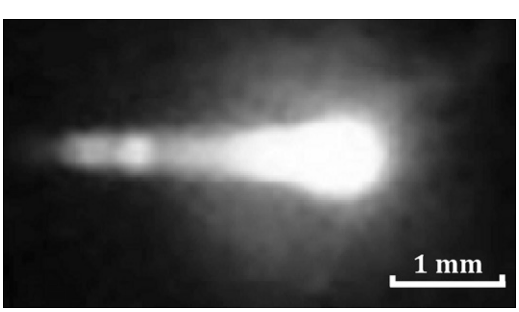

Fig. 3. Plasma image obtained during the breakdown in acetone (E_{laser} = 100 mJ, $P_{acetone}$ = 220 mbar).

For acetone, the plasma is purple and is less bright. For mixtures of these gases, the color has an intermediate behavior. Fig. 4 shows the time evolution of light intensity during a breakdown. During the first stages of breakdown, the light intensity increases very rapidly. The laser beam provides energy to the plasma, which leads to the increase of its size and temperature. At the end of the pulse, this growth stops. The light emission begins to decrease, because energy is dispersed into the surrounding environment. The lifetime of the plasma is about 3.5 ms, which is much longer than the duration of laser emission (about 5 ns).

3.2. Breakdown probability and energy absorption

Fig. 5 shows the probability of breakdown in air (P = 1 bar), in acetone/air mixtures (P = 1 bar) for equivalence ratios equal to 1, 1.4 and 2 and in pure acetone (P = 220 mbar). For all mixtures, there is a threshold energy that allows the breakdown (Table 2). Below this value, it is not possible to create a plasma. Above this value (E_1), the breakdown probability increases with incident energy until it reaches 100%. However, the shape of the curves changes depending on gas composition. The difference between E_1 and E_{100} is indeed higher when the percentage of air in the mixture increases. In addition, obtaining a breakdown requires less energy in a mixture

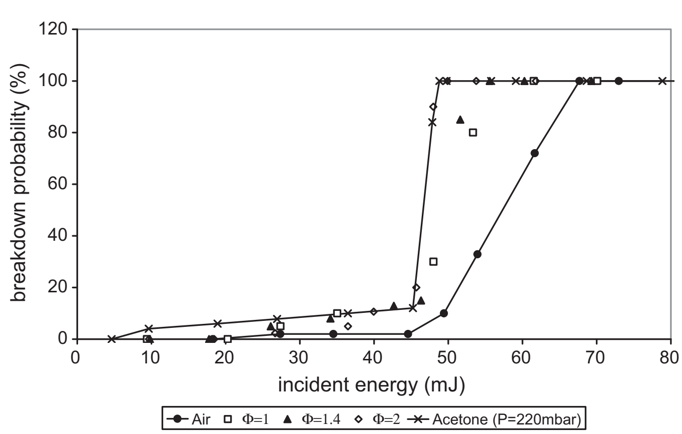

Fig. 5. Breakdown probabilities.

Table 2
Breakdown thresholds.

	Air (P = 1 bar)	Acetone/air mixture (P = 1 bar)			Acetone (P = 220 mbar)
		Φ = 1	Φ = 1.4	Φ = 2	
Thresholds (mJ)					
E_1	22.8	21.7	19.4	19.3	5.9
E_{50}	58.9	50.1	49.0	46.0	46.6
E_{100}	67.5	61.5	55.6	49.9	48.8

containing a large amount of acetone. Acetone therefore promotes the breakdown. This is not surprising since acetone has an ionization potential of 9.7 eV, which is lower than that of air (15.6 eV).

From the data measured by the energy meters, the energy absorbed during the experiments (with and without breakdown) was calculated for different compositions. The attenuation of the laser energy in the presence of breakdown is attributed to spark absorption and to the scatter of the laser light by the developing spark of plasma. However, as discussed by Ma et al. [24], losses due to diffraction are negligible and the attenuation can be considered to be due solely to the absorption by the spark. Therefore, the energy absorbed by the plasma (E_{abs}) is equal to the difference between the energy entering in the chamber (incident energy noted E_i) and the transmitted energy through the plasma (noted E_{tr}):

$$E_{abs} = E_i - E_{tr} \quad (3)$$

The incident energy was obtained from the measurement of the energy meter located just after the laser (2a in Fig. 1) by considering the absorption of the lens and of the first window of the chamber. The transmitted energy is given by the energy meter placed after the chamber (2b in Fig. 1) taking into account the absorption of the second window of the chamber.

Fig. 6 presents the measurements in air, in a stoichiometric mixture of acetone/air and in acetone. The shape of the curve corresponds to those available in the literature [11,15]. Three areas are visible:

- Below E_1, there is no absorption of the beam as no breakdown occurs.
- Between E_1 and E_{100}, a transition zone appears. Some shots involve a breakdown while others do not. When the breakdown occurs, energy is absorbed by the plasma, which corresponds to the points above the x-axis.
- Above E_{100}, the breakdown is systematic. All shots lead to breakdown and to energy absorption.

For all the mixtures tested, there is great variability of the absorbed energy for a given laser energy. This is even more pronounced when the mixture contains only acetone. This variability in the energy deposit is due to the stochastic aspect of the laser-induced breakdown phenomena.

3.3. Ignition probability

Fig. 7 shows the ignition probability of acetone/air mixture for equivalence ratios equal to 1.2 and 2.4. The curves for 1.2 are typical results obtained for mixtures within the flammable range while those of 2.4 shows the behavior at flammability limits. Within the flammable range, the probabilities of breakdown and of ignition increase when the incident energy increases. However, a breakdown does not automatically lead the mixture ignition. Energy absorbed by the plasma may be too low to cause ignition. The probability of ignition may be below those of breakdown. In the case of experiments at the flammability limits, the probability of ignition does not increase necessarily with increasing incident energy. This

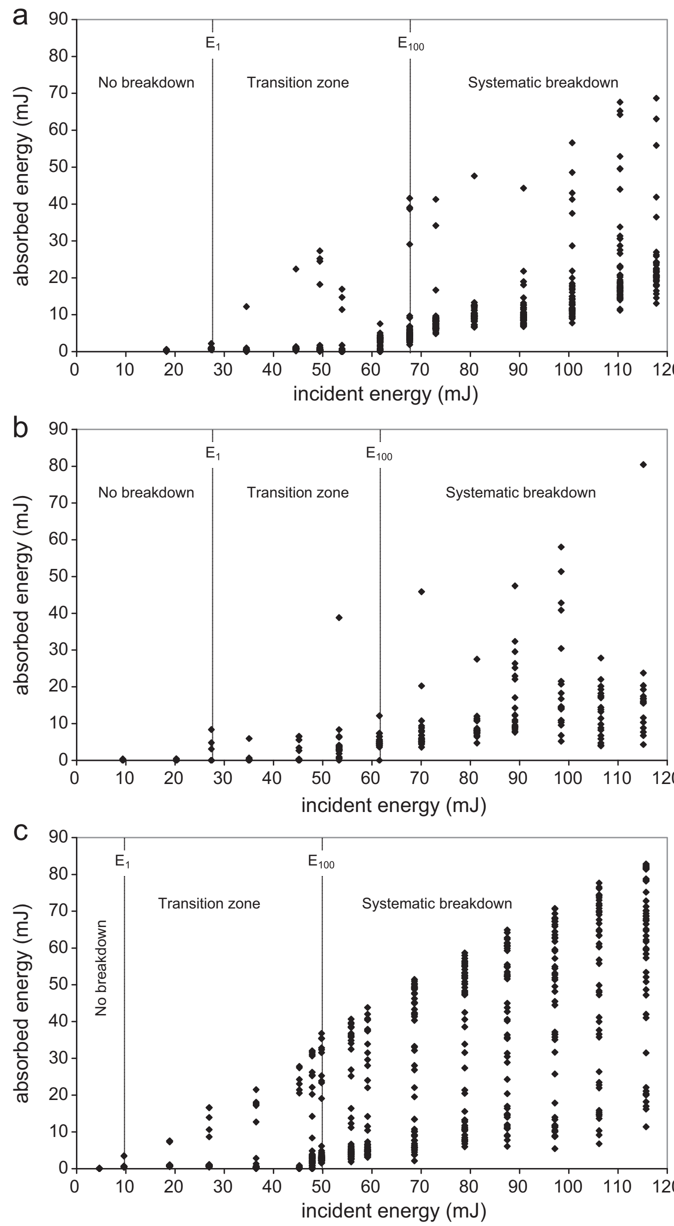

Fig. 6. Absorbed energy versus incident energy for: (a) air (P = 1 bar); (b) a stoichiometric mixture of acetone/air (P = 1 bar) and (c) acetone (P = 220 mbar).

is due to the variability of deposited energy for same laser energies (Fig. 6). In most studies of laser-induced ignition, the probabilities of ignition are expressed according to the energy delivered by the laser [23,30]. Given the obtained results, it seemed more appropriate to express the probability of ignition versus energy absorbed by the plasma during breakdown. Fig. 8 presents the results for all equivalence ratios. For equivalence ratios between 1.4 and 1.8, little energy is needed to ignite the mixture. 100% of ignition is indeed obtained when the absorbed energy is greater than 5 mJ. For equivalence ratios equal to 1.2 and 2, a reliable ignition is obtained with absorbed energies greater than 10 mJ. For an equivalence ratio of 2.2, it is necessary to provide at least 18 mJ for having 100% of ignition. For equivalence ratios of 1 and 2.4, we failed to obtain a reliable ignition during our experiments. The maximum probability, that we were able to record, was 60 and 85% for equivalence ratios of 1 and 2.4, respectively. In addition to energy changes, we note that the equivalence ratio also has a role on the shape of the curve. For equivalence ratios between 1.4 and 1.8, the energy gap between an

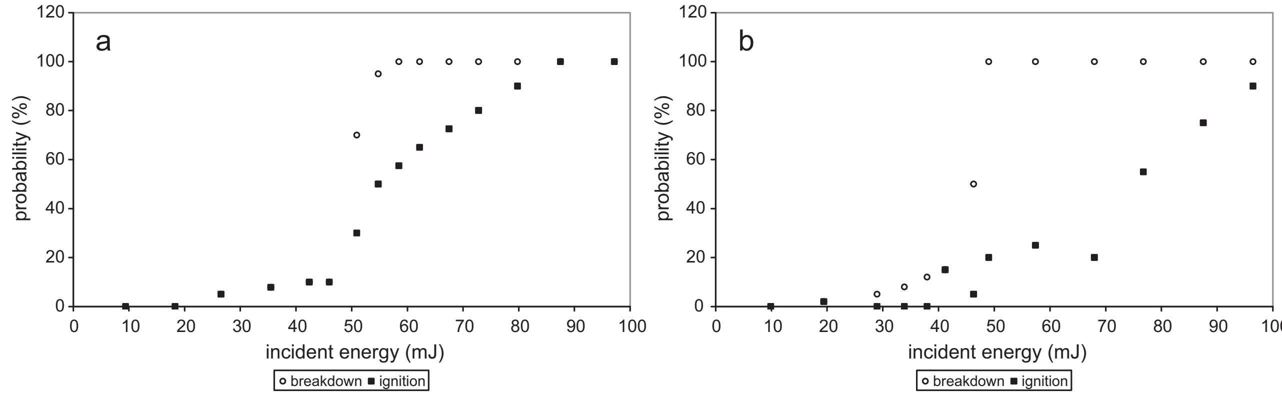

Fig. 7. Probabilities of breakdown and of ignition for acetone–air for an equivalence ratio equal to: (a) 1.2 and (b) 2.4.

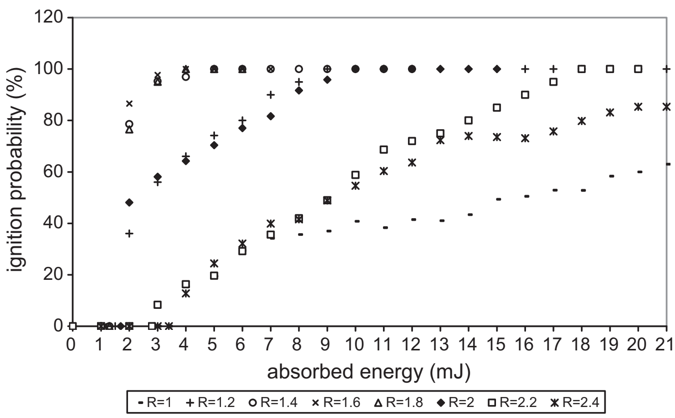

Fig. 8. Ignition probability versus absorbed energy for all equivalence ratios.

ignition probability of 0 and 100% is thin. When equivalence ratios deviate from these values, the gap becomes increasingly large. It is therefore more interesting to use a mixture with an equivalence ratio to ensure the ignition and the reproducibility of the phenomenon.

3.4. Minimum ignition energy (MIE)

In the hazard assessment, it is more appropriate to focus on the minimum ignition energies (MIE) rather than on the certain ignitions. As explained previously, MIE were determined by using the approach of Moorhouse [3]. Fig. 9 and Table 3 present the results. The minimum ignition energies remain at their lower values for mixtures having an equivalence ratio between 1.4 and 1.8. Then they increase sharply to about 10 mJ and 3 mJ for equivalence ratios of 0.9 and 2.4, respectively. Increases in the minimum ignition to both the lean side and the rich side of the stoichiometry are similar to the studies reported in literature for different mixtures [21,25,29,33]. The U-shaped curve has is minimum for an equivalence ratio of 1.6 and an absorbed energy of 1.15 mJ. This MIE corresponds to the value measured with electrical sparks [2,34]. This is quite surprising because usually the minimum ignition energies obtained with a laser-spark ignition are greater than those measured by electrical sparks [20,35].

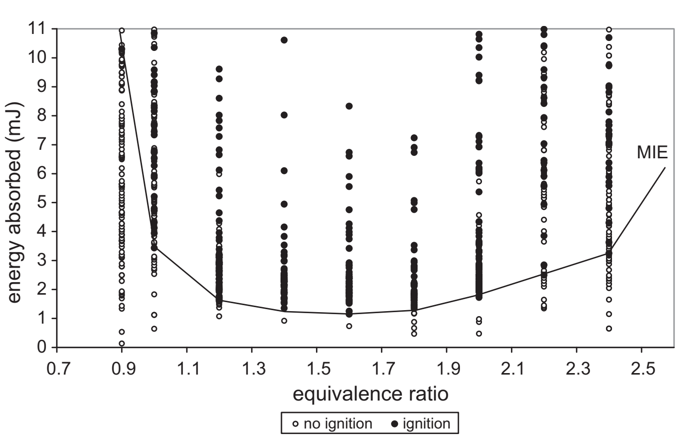

Fig. 9. Laser ignition diagram for acetone at 22 °C and 1 bar.

3.5. Kernel temperature at the minimum ignition energy

To characterize in more detail the plasma, the kernel temperature was calculated from the absorption coefficient of the spark for each minimum ignition energy.

The absorption coefficients were calculated using the Beer–Lambert relation:

$$\frac{E_{tr}}{E_i} = \exp(-K_v l) \tag{4}$$

where K_v is the absorption coefficient and l is the length of the focal volume equal to 683 μm (Eq. (2)).

The absorption coefficient of each spark causing the ignition with the minimum energy is given in Table 3. They are between 0.39 and 2.74 cm^{-1}. Phuoc and White [25] showed that the absorption coefficient depended on the incident energy and ranged between 0.1 and 100 cm^{-1}. These values are consistent with our results. In our study, the minimum ignition energy occurs indeed for a laser beam with an energy between 27 and 60 mJ, corresponding to low values of K_v reported by Phuoc and White [25].

If it is assumed that the absorption of the laser energy by the spark is primarily due to the electron-ion inverse Bremsstrahlung process, in which light is absorbed as a result of free–free transitions of the electrons in the field of the ions, the kernel temperature can be estimated with the information on K_v. It has been reported that when air is approximately 1% ionized, the effective absorption coefficient for inverse Bremsstrahlung can be calculated using the following equation [24,25]:

$$K_v = \left[1 - \exp\left(-\frac{hc}{\lambda kT}\right)\right]\left(\frac{4e^6\lambda^3}{3hc^4 m_e}\right)\left(\frac{2\pi}{3m_e kT}\right)^{1/2} n_e \sum z_i^2 n_i g_i \tag{5}$$

where n_e is the electron number density (cm^{-3}), λ is the wavelength (cm), k is Boltzmann's constant (1.3803×10^{-16} erg/K), h is Planck's constant (6.6237×10^{-27} erg s), c is the speed of light (2.9979×10^{10} cm/s), m_e is the electron mass (9.109×10^{-28} g), e is the electronic charge (4.8029×10^{-10} abs esu), T is the temperature

Table 3
Minimum ignition energies, corresponding absorption coefficient and kernel temperature for acetone/air mixtures at 1 bar.

Equivalence ratio	0.9	1	1.2	1.4	1.6	1.8	2	2.2	2.4
MIE (mJ)	10.30	3.42	1.60	1.36	1.15	1.34	1.72	2.50	3.25
K_v (cm^{-1})	2.74	1.16	0.47	0.39	0.42	1.12	0.97	1.51	1.20
T ($\times 10^6$ K)	0.9	1.5	2.8	3.2	3.0	1.6	1.7	1.3	1.5

(K), z_i is the charge of the ith ionic species, n_i is the number density of the ith ionic species (cm^{-3}) and g_i is the Gaunt factor.

In order to facilitate calculation of absorption coefficients, a simplified form has been used based on Kramer's formula [24]. Substituting values of all parameters in standard electrostatic units (e.s.u.), as listed above and assuming, $\sum z_i^2 n_i g_i = n_e$, one obtains Kramer's formula:

$$K_v = 3.69 \times 10^8 \frac{n_e^2 \lambda^3}{T^{1/2} c^3} \left[1 - \exp\left(-\frac{hc}{\lambda kT}\right)\right] \qquad (6)$$

For this study, Eq. (6) becomes:

$$K_v = 1.65 \times 10^{-35} \frac{n_e^2}{T^{1/2}} \left[1 - \exp\left(-\frac{13521}{T}\right)\right] \qquad (7)$$

To calculate the kernel temperature, the electron density must be known. For a laser operating at 1064 nm, the electron density can be assumed equal to 10^{20} cm^{-3} [24,25,36]. Using the absorption coefficients in Table 3, the temperature kernels were calculated. The values are ranged between 9×10^5 and 3.2×10^6 K (Table 2) with a maximum temperature near the equivalence ratio corresponding to the smallest MIE. These results are in agreement with the reported kernel temperatures around 10^6 K by Ma et al. [24] and Phuoc and White [25] for small focal length.

4. Conclusions

The breakdown and the laser-induced spark ignition of acetone–air mixtures were experimentally investigated. The results can be summarized as follows:

- The breakdown probabilities vary according to the composition of the gas mixture. The presence of acetone in air tends to reduce the energy required to obtain a breakdown. This is likely due to the ionization energy of acetone which is lower than that of air.
- The breakdown in the mixture leads to the formation of a plasma which induces an energy absorption. However, for a given incident energy, there is a wide variation in the energy absorbed by the plasma. This result prompted us to characterize the ignition properties according to the energy absorbed by the plasma instead of the energy sent by the laser, unlike the studies usually done with lasers.
- The ignition probabilities and the minimum ignition energies were determined in the equivalence ratio range 0.9–2.4. The results show an increase of ignition energy to the lean and rich side of the stoichiometry. The minimum ignition energy was obtained for an equivalence ratio of 1.6 and an absorbed energy of 1.15 mJ, which corresponds to value recorded with electrical spark ignition.
- The absorption coefficient and the kernel temperature of the plasma leading to minimum ignition energies were calculated. The absorption coefficients are between 0.39 and 2.74 cm^{-1} what corresponds to kernel temperatures between 9×10^5 and 3.2×10^6 K.

Thus, this work provides improvements in understanding the laser-induced spark ignition in acetone–air, which could be useful for risk assessments.

Acknowledgement

The authors express their gratitude to François Vaslin for his help on the finalization of the experimental device.

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