Abstract

The first stages of laser-induced spark ignition were investigated as a function of time. Experiments were conducted using a premixed laminar CH$_4$/air burner. Laser-induced breakdown was achieved by focusing a 532-nm nanosecond pulse from a Q-switched Nd-YAG laser. An anti-reflection-coated lens with a focal length of 100 mm was used. The results obtained from an intensified high-speed and PIV CCD camera and a Cassegrain optics system coupled to an ICCD spectrometer provided information about the formation of laser-induced plasma and its transition to a flame kernel and a self-sustaining flame. The localization of the kernel and its time development were reproducible. Two types of flame fronts develop: one that expands against the flow direction, and one that moves with the flow. The initial flame expansion along the laser axis is asymmetric due to the shape of the plasma, different ionization levels inside the plasma, and the shock-wave expansion. Development of the fast flame occurs behind the shock wave induced by the plasma. This is important when laser ignition is used as a flame holder. An ICCD spectrometer coupled to an optical fiber permitted chemiluminescence visualization. The spectrum obtained during the plasma and flame kernel formation defined different stages in flame formation. The results obtained with these two optical techniques were synchronized to obtain the temporal resolution of the flame kernel evolution. Laser induced ignition of a very lean mixture can be controlled to provide local heat release and extinction in a flame.
Introduction

The ignition of a lean mixture is important as it affects pollutant emission and engine consumption. Ignition is also an important design factor for gas turbines and rocket combustors. Therefore, there has been much research to increase our knowledge of laser-induced ignition [1-9]. There are numerous advantages to laser-induced ignition compared to electrical spark ignition: the amount of energy and the rate of its deposition can be controlled, the timing can be adjusted, and the optimal ignition location can be chosen. Furthermore, multi-point ignition can be used without losing heat during the flame development because of a material surface near the flame kernel. Another application of laser-ignition is spark-flame holding. This prevents the stagnation pressure losses that occur behind a solid flame holder and eliminates the screech phenomenon [10, 11]. However, the implementation of this technology has been limited by our understanding of the details of ignition, such as the parameters that influence laser-induced breakdown and flame generation. The aim of this research is to improve our understanding of the earliest stages of flame formation. It is well known that when a laser beam of the order $10^8$ W/cm$^2$ interacts with gas, high temperature and pressure plasma are generated [12, 13]. Four mechanisms exist to produce a laser-induced spark [11], depending essentially on the laser wavelength and the mode of energy deposition: thermal initiation [14], non-resonant breakdown, resonant breakdown, and photochemical ignition [15].

We investigated laser spark ignition experimentally using a nanosecond pulse from a Q-switched Nd:YAG laser. This laser beam is typically used to obtain cascade electrons generated by multi-photon ionization [12]. A spark produced in this manner has a smaller time scale than the kinetic energy time scales and the chemical induction time. This generates a rapidly expanding shock wave that can be of sufficient strength to ignite a gaseous combustible mixture. The strength and duration of the generated shock wave are important to flame ignition and development. The measurements obtained from this work yielded high spatial and temporal resolution of plasma evolution, from its formation to the initiation of a self-sustaining flame.

Experimental setup

Figure 1 shows a schematic of the experimental apparatus, which can be separated into three main parts: the ignition laser, the acquisition equipment, and the synchronization system.

![Fig. 1. Experimental set-up](image)

1. Nd:YAG Laser
2. Laser power unit
3. Beam splitter (5%)
4. Laser attenuator
5. Lens
6. Energy meter
7. ICCD spectrometer
8. Computer
9. Delay generator
10. Cassegrain optics
11. CCD camera
12. Energy meter display
Laser-induced breakdown was achieved by using an anti-reflection coated lens to focus the output of the second order harmonic of a double-cavity Nd:YAG laser (Spectra Physics PIV-400) operating as a Q-switched nanosecond (ns) laser. The laser delivered pulse widths of 8 ns at 532 nm. Spark energy measurements were made by splitting off 5% of the laser pulse and comparing it with the energy transmitted after the breakdown. Syage et al. [1] and Ma et al. [2] attributed the attenuation of laser energy that occurs when a spark forms to laser-induced breakdown. They observed negligible diffraction effects. The reference and transmitted pulse energies were measured using two laser precision pyroelectric energy meters (Ophir: PE-25). The breakdown threshold ($E_{\text{mini}}$) in this experimental configuration was 10 mJ/pulse. The experiments were run for 1.5 $E_{\text{mini}}$.

The plasma formation images obtained by focusing the laser were acquired by a CCD camera (TSI-Model 630046 PIVCAM 10.30) coupled to an intensifier (Hamamatsu: C6654). This system allowed image acquisition with an integration time of 50 ns. Images of the flame kernel expansion were obtained with a high-speed CCD camera (Phantom: V5.0 SCR-CMOS tech) coupled to an intensifier (Hamamatsu: C6654), which allowed a frame rate of 1000 pictures per second (pps) at full resolution (1024*1024 pixel$^2$) and 60,000 pps at lowest resolution (256*32 pixel$^2$). Emission spectra of the spark were obtained from chemiluminescence by using an ICCD-spectrometer (Andor: Oriel MS-257) coupled to Cassegrain optics [16, 17]. The Cassegrain optics system, shown in Figure 2a, gave high spatial resolution by minimizing the spherical aberration of the pair of mirrors and by avoiding chromatic aberration. A Cassegrain optics system has spatial and temporal resolution that is higher than, for example, that of single-lens optics coupled to an optical fiber [18]. The observation volume of the Cassegrain optics was estimated to be 100 µm in diameter and 0.8 mm in length (Fig. 2b) from the relative intensity (threshold level as $e^{-2}$ times the peak value) [19]. The Cassegrain optics were placed on a traverse system (ITO: accuracy: 0.5 µm) to allow three-dimensional displacement. The collected emissions were guided to the ICCD spectrometer through a multi-optical fiber (core: 200 µm).

The timing system supplied TTL pulses that were obtained from a Stanford Research System DG 535-X. The experiments were performed in single laser shot mode to avoid perturbations produced by previous spark events. The delay between laser pulse and acquisition could be adjusted independently to obtain measurements at different times after the laser-induced spark. The time resolution was estimated to be 2 ns. The horizontal and vertical resolutions for image acquisition were each approximately 40 µm (field of view ($F_v$) = $4\times4$ cm$^2$) for the high-speed camera system and 4 µm ($F_v$ = $4\times4$ mm$^2$) for the ICCD system.

Two flow controllers (STEC: accuracy: 0.1 l/min for air and 0.01 l/min for CH$_4$) were used to regulate the flow of methane and air. Gases were mixed in a chamber at atmospheric pressure and then directed to an atmospheric pressure laminar Bunsen burner (diameter = 8 mm). The mixture velocity at the burner exit was 1.15 m/s for an equivalence ratio of 0.9. The mixture was at atmospheric pressure and temperature before ignition. The laser beam was focused 2.5 cm from the burner exit.

Results and discussion

Two sets of visualization equipment were used to observe the temporal development, because of the time scale differences between plasma formation and flame kernel development. ICCD cameras have been used for plasma development; they give a temporal resolution of 1 ns and offer good reproducibility
between two sparks at the same level of energy [1, 4]. Therefore, an ICCD camera was used to obtain images for different time events. A high-speed camera was used to visualize the temporal evolution of the flame kernel. Images obtained with this camera were recorded by synchronizing the experiment to the nearest nanosecond.

The ignition process observed with the ICCD camera is described first. Figures 3a-c show three typical images obtained 100, 200, and 1000 ns after the laser pulse. The integration time was fixed at 50 ns.

![Figure 3a-c. Time series of plasma to flame kernel development, premixed CH₄/air flow, equivalence ratio=0.9, flow velocity= 1.15 cm.s⁻¹. Eₚulse= 15.4 mJ. (a) integration time (tᵢ)= 50 ns, intensification (Iᵢnt)= 0 (b) tᵢ=50 ns, Iᵢnt=4 (c) tᵢ = 450 ns, Iᵢnt= 9. The spark occurs 2.5 cm from the burner’s exit.](image)

Figure 3a shows the plasma shape 100 ns after the laser pulse; it is cylindrical, with varying intensity along the x-axis but symmetrical along the y-axis. The differences in intensity along the plasma main axis are due to two physical processes: the energy absorption mode during the plasma formation (Inverse Bremsstrahlung process) and the multi-point ignition resulting from spherical aberrations caused by the plano-convex glass lenses [12]. These aberrations deform the laser beam shape from Gaussian to one with different maximum energy positions along the laser path. Figure 4 shows typical spectra obtained 100 ns after the laser pulse.

![Figure 4. Plasma spectra, f= 100 mm, 100 ns after laser pulse, Eₚulse= 15 mJ. Average of 50 spectrums with an integration time of 10 ns. tᵢ=10 ns, Iᵢnt= 5 for X=0 and X=0.5 mm, Iᵢnt= 3 for X=-0.5 mm](image)
The initial localization \((x = 0)\) was defined as the location of the generation of the first electron, and corresponds to the first signal observed by the camera and the ICCD spectrometer 1 ns after the laser pulse. The accuracy of this localization can be roughly estimated as half the diameter of the measurement probe volume (i.e., 0.05 mm). Negative values correspond to locations closer to the laser. There was a high level of ionization in the range of 200 to 600 nm, characterized by a continuous background. The ionization was higher at \(x = -0.5\) mm due to laser light absorption across a short distance. At \(x = -0.5\) mm, the intensifier gain was fixed at 3 due to the higher intensity; at other positions, the gain was fixed at 5. The wavelength characteristic of the combustion species could not be observed, since the time scale was too small compared to the chemical reaction time.

Figure 3b shows the shape of the plasma observed 200 ns after the laser pulse. The size of the plasma has increased due to the ionization caused by the plasma itself. Spectra corresponding to this picture (Fig. 5) show a more homogeneous ionization inside the plasma and the appearance of species at specific wavelengths (C, N, N\(^+\), O, etc.) 200 ns after the laser pulse. It was not possible to observe the characteristics of some of the combustion radicals due to the time scale.

![Graph showing plasma spectra](image)

**Fig. 5.** Plasma spectra, \(f = 100\) mm 200 ns after laser pulse, \(E_{\text{pulse}} = 15\) mJ. Average of 50 spectra with an integration time of 10 ns, \(t_i = 10\) ns, \(I_{\text{int}} = 5\) for \(X=0\) and \(X=0.5\) mm, \(I_{\text{int}} = 3\) for \(X=-0.5\) mm

Figure 3c is an image obtained 1 \(\mu\)s after the laser pulse, showing two “bowls” of intensity located on the laser path at the extremes of the plasma. The formation of these bowls occurred roughly 1 \(\mu\)s after the laser pulse, and they were visible for 4 \(\mu\)s. The bowls were not observed during laser-breakdown in the air, so they are presumed to be correlated with the combustion phenomenon. Images obtained after 5 \(\mu\)s do not show this type of behavior, indicating that it was a local event that did not lead to flame propagation. Figure 6 presents the spectra obtained at the center of a bowl 200 ns, 1 \(\mu\)s, and 2 \(\mu\)s after the laser pulse. Before the separation of the two bowls (1 \(\mu\)s after the laser pulse), the spectrum indicates a high ionization level with an important continuum background. After 1 and 2 \(\mu\)s, the spectra show more distinct lines that correspond to ionized atoms, but it is not easy to observe some of the radicals usually observed in flames (OH\(^-\), CH\(^+\), C\(_2\)\(^+\), and so forth).
Fig. 6. Flame kernel spectra (X= +1mm), f= 100 mm, 200 ns, 1µs and 2µs after laser pulse, E_{pulse}= 15.4 mJ. t_i= 10ns, I_{int}= 5. The spark occurs 2.5 cm from the burner’s exit. Average of 50 spectra with an integration time of 10 ns.

Figures 7a-j are a series of images obtained with the high-speed camera during the development of the flame. These were obtained with a 1,000-image per second frame rate for the methane/air mixture with an equivalence ratio of 0.9. A rapidly-expanding flame developed behind the shock wave produced by the plasma (Figs. 7a-d). The location of the center of the flame kernel was stable over roughly 5 ms, which demonstrates that the flow had a low impact on kernel formation due to the time-scale difference between the formation of the shock wave and the flow velocity. The flame was toroidal in shape, as observed elsewhere [4, 10]. The development of a toroidal flame kernel is a universal phenomenon of spark ignition [4, 20, 21]. Furthermore, this phenomenon is dependent only on the discharge shock wave and not on the gas composition. Thus, the toroidal shape is dependent on the spark energy. After 3 ms (Figs. 7c-j), the flame began to develop asymmetrically against the flow. The development of the flame kernel was dependent on the flow during this period. The development against the flow halted after 8 ms (Fig. 7d), while the fast flame continued to expand in the flow direction. The reverse flow flame remained at the same location for about 4 ms (Figs. 7d-e) before moving in the flow direction. During this time, the shape of the flame-front changed, due to the modification of flame velocity direction and the growth of the flame-front along the radial axis. The radial expansion of the flame reached a diameter greater than 8 mm, which signifies that the equivalence ratio at this location was less than 0.9. The flame deceleration was in part due to the CH_4/air flow, and in part due to the transition from a spherical to a planar flame front. In Figs. 7h-j, the flame propagates with the flow at a constant velocity.

Additional information about the development of the flame kernel can be obtained from this series of images. First, the plasma generates a rapidly expanding shock wave of sufficient strength to ignite the CH_4/air mixture. The flame kernel is an asymmetric toroidal shape due to the expansion mode of the shock-wave. The asymmetric behavior can be attributed in part to the plasma characteristics (shape and ionization level). Different ionization levels in the plasma are visible in Figure 4, 100 ns after the laser pulse. Figure 8 shows the typical plasma shape; this image was obtained with a lens of 100 mm focal length, 200 ns after the laser onset in the CH_4/air mixture. There is a similarity between the asymmetric shapes of the plasma and the flame kernel, caused by the energy deposition in the plasma, which is a multi-photon ionization followed by a cascade growth. Most of the pulse energy is absorbed in the laser side of the plasma. Therefore, the plasma shows the dissymmetry of energy between the laser-side and the opposite side of the plasma, which explains in part the backwards flame propagation along the path of the ignition laser beam.
The initial flame expansion velocity before 0.5 ms was heavily correlated to the spark energy. Afterwards, the velocity depended mainly on the equivalence ratio. This is in good agreement with Spiglanin et al. [4]. Using a simple Taylor blast-wave model to describe the shock-wave characteristics [22], these authors observed some minor differences in calculations performed using stoichiometric and lean mixtures. The velocity of the first flame decreased rapidly with distance. This can be correlated to the rapidly decreasing shock-wave velocity and to the weak shock observed by Phuoc et al. for a laser-induced spark in air [23]. The flame velocity during the first 1 ms was estimated from high-speed camera visualizations to be higher than 50 m/s. This velocity decreased rapidly when moving away from the plasma location. After roughly 1 mm, the flame velocity reached a value close to 2 or 3 m/s, depending on the mixture equivalence ratio. Therefore, the velocity behind the shock wave was independent of the equivalence ratio during the first stage of flame expansion, but became dependent after a distance of 1 mm. This distance is a function of the decrease in the shock wave velocity, which is correlated to the spark energy. Ignition failure can occur at low energies due to this phenomenon, because the flame kernel must reach a certain diameter to propagate, similar to an electrical spark [24, 25]. Experiments with a small amount of energy deposition showed small flame-kernel generation, which resulted in ignition failure, as observed by Spiglanin et al. [4].
Several images obtained with the high-speed camera showed a curved halo at the top of the picture. Its position was stable, but its size and intensity changed. No direct link between the halo and flame front could be defined, and it appeared only during flame formation.

**Conclusion**

This work presents time-resolved images of an expanding flame kernel generated by laser-induced breakdown. The results allow the flame ignition in a laminar flow to be divided into several steps, corresponding to different time scales.

\[ T < t_o + 1 \mu s \]  

This period corresponds to plasma formation and cooling. Complete ionization is observed in the plasma, followed by an expansion during the Bremsstrahlung process. The time scale of the phenomena is 10-100 ns. Shock-wave formation and detachment from the plasma occur during this period (Phuoc et al. [23]).

\[ t_o + 1 < T < t_o + 100 \mu s \]  

During this period, two high-intensity bowls form at the plasma extremes along the laser path. The formation of these bowls can be attributed to an energy exchange from the plasma kernel to the surroundings due to low shock-wave expansion near the plasma extremes. The time scale of the phenomena is of order 1 \( \mu \)s. This stage corresponds to “ignition starting.”

\[ t_o + 100 < T \mu s \]  

During this stage, flame ignition is observed behind the path of the shock wave. The flame kernel is a toroidal asymmetric shape, caused in part by the shock-wave expansion mode (opposing gas flows colliding in the center of the flame kernel) and in part by the dissymmetric energy repartition in the plasma due to the energy absorption mode.

The flame velocity in the first stages of kernel formation is directly correlated to the spark energy, and becomes dependent on the equivalence ratio after a distance that depends on the shock-wave expansion (spark energy). This dependence leads to the existence of a critical radius to sustain flame propagation, similar to an electrical spark.
References


[23] Phuoc X.T. and White C.M., Optical characterization of the laser induced spark in air, Optical diagnostics in engineering, Vol. 5, Part. 1, ISSN: 1364 (2001)
