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S. H. Chan
Department of Mechanical Engineering
University of Wisconsin-Milwaukee
Milwaukee, Wisconsin USA

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DETAILED CHARACTERIZATION OF MINIMUM IGNITION ENERGIES OF COMBUSTIBLE GASES USING LASER IGNITION SOURCES

Elliot H. Lim*, Andrew McIlroy†, Paul D. Ronney* and Jack A. Syage+

*Department of Mechanical Engineering
University of Southern California
Los Angeles, CA 90089-1453

†Mechanics and Propulsion Department
The Aerospace Corporation
Los Angeles, CA 90009-2957

ABSTRACT

Lewis and von Elbe's data on minimum spark ignition energies in gases have been the standard for many years, however, these data still do not agree with the most detailed computational models available. With this motivation, their classic experiment was revisited using laser ignition sources, with an emphasis on better characterization of the ignition source and its effect on minimum ignition energy. The apparatus consisted of a laser ignition source operating either as a Q-switched nanosecond or a pulse mode-locked picosecond laser. For CH₄-air mixtures of varying stoichiometry the minimum ignition energy was bracketed through repeated trials at varying laser spark energies. Laser spark kernel sizes were quantified by imaging the visible emission of these sparks. Results showed that the laser ignition experiments are consistent with Lewis and von Elbe's measurements for lean and rich mixtures, however, for near-stoichiometric mixtures, the laser ignition values were higher. These results are interpreted in the context of the size of the energy deposition region. Further studies to assess this interpretation are identified.

INTRODUCTION

The study of ignition of premixed combustible gases is of great fundamental and practical importance for internal combustion engines and gas turbines as well as fire and explosion hazard assessment. For many years, the pioneering work of Lewis and von Elbe [1] has provided the baseline data on the minimum ignition energy (MIE) required to produce flame propagation. Using electric sparks, the effects of mixture composition, pressure, spark gap, etc. on MIE was measured. However, these and other experimental data do not agree with computational predictions. For example, Lewis and von Elbe reported an experimental value of MIE of 0.4 mJ for a stoichiometric CH₄-air mixture at 1 atm while the computational prediction of Sloane and Ronney [2] using detailed chemical, hydrodynamic, and transport models, is 0.10 mJ. Also, Lewis and von Elbe infer that the minimum flame kernel diameter is about 2 mm, based on the spark gap providing the lowest MIE, whereas Sloane and Ronney predict 0.6 mm. Also, while Lewis and von Elbe clearly recognized the importance of heat losses to the spark electrodes, they did not report the diameter of the electrodes they used. These discrepancies between experiments and computational prediction so far have been unresolved. However, a more recent work by Kingdon et al. [3] has shown that the laser ignition deposition, a role in ignition energies can be more accurately measured in practical situations. The advantages of using laser ignition sources in actual engines, as opposed to spark ignition sources, is demonstrated in this study. The laser ignition source was characterized in terms of heat deposition and its effect on the ignition process. The results are in agreement with the computational predictions and provide a better understanding of the ignition process.
and computations may possibly be due to an inadequate characterization of the ignition source, given the limitations of the instruments available at the time of Lewis and von Elbe's experiments. Alternatively, aspects of the physics of energy deposition, not present in even the most detailed models available to date, may play a role in ignition efficiency. For example, the MIE values for laser ignition of H₂-air mixtures measured by Syage et al [3] exceed the electric discharge measurements by a factor of 6 at stoichiometry. Toward the lean and rich limits, the discrepancy narrows but does not disappear.

Consequently, the goal of this work is to employ modern lasers and optical instruments to produce ignition sources, characterize these sources and compare measured ignition parameters to previous experiments and computational models. Kingdon and Weinberg [4] conducted a study along these lines over 20 years ago, but, as discussed below and in more detail by Ronney [5], the applicability of their results is affected by the size of the energy deposition region in their experiments, and the fact that to facilitate breakdown they employed thin wires of varying materials as laser targets. The critical role of the size of the ignition kernel on MIE was first shown experimentally by Lewis and von Elbe [1] and computationally by Frendi and Sibulkin [6].

The laser-based techniques to be employed here has several likely advantages over the classical minimum ignition energy experiments. First, the actual energy deposited in the gas, rather than just the energy stored in a capacitor bank, is measured. Second, there are no electrodes to act as heat sinks. Third, the distribution of energy within the ignition region is readily characterized by optical means. For these reasons, it is believed that this system can provide a more accurate and complete characterization of the ignition event. If the mechanism of energy deposition has bearing on MIE, then one might observe different MIE values by different ignition sources. A collection of results would form a base of knowledge that would assist in developing more refined models of flame ignition and may provide insight into the long-standing discrepancies between electric spark experiments and computational predictions.

Of course, there are substantial differences between different types of laser ignition sources and electric sparks. A detailed discussion of these differences is given by Grey Morgan [7]. Here we note that many of the differences between MIEs for laser and electric spark ignition sources can be understood based on relatively simple thermal models of ignition [5]. An important distinction is that for laser sources, the energy is deposited mostly in a central core region whereas for electric sparks, the cathode and anode voltage drops cause a large portion of the energy to be deposited near the ends of the deposition region. The potential impact of the some of the differences between laser ignition sources and electric sparks is addressed in the Discussion section.

For laser sources, the breakdown process generally begins with multiphoton ionization of a few gas molecules, which release electrons that can then readily absorb more photons, increasing their kinetic energy. The electrons liberated by this means collide with other molecules and ionize them, leading to an electron avalanche and breakdown of the gas. Usually multiphoton processes are essential for the initial stage of breakdown because the available photon energy at visible and near-UV wavelengths is much smaller than the ionization potential of most molecules. For very short pulse durations (typically a few picoseconds) the multiphoton process alone must provide breakdown, since there is insufficient time for electron-molecule collisions to occur. Of course, there are loss processes (such as by diffusion of electrons out of the focal volume, radiation, collisional quenching of excited states, etc.), which render the critical intensity for breakdown dependent on the focal volume, pressure, type of gas, impurities, etc. Because laser spark breakdown consists of multiphoton absorptions in the presence of losses, the
optical intensities needed to induce breakdown in gases are extraordinarily high. This indicates the need for high-energy laser sources with short pulse durations and tightly focused beams so that the pulse energy can be sufficiently concentrated in space and in time to produce breakdown. These conditions have led to difficulties in producing pulses of energy which are smaller than the MIE in many investigations, especially for highly reactive mixtures with low MIE. These issues are given special consideration in the design of the experiments described below.

EXPERIMENTAL

A schematic of the experimental apparatus is presented in Fig. 1. The reaction chamber consisted of a 4-in. Conflat cube that was Teflon coated to eliminate metal surface interactions [3]. As a safeguard, the reaction chamber was connected to a 5 liter evacuated buffer chamber separated by an Al foil membrane. A successful ignition generated a pressure rise which ruptured the Al foil. Two lenses served as the entrance and exit windows for the laser pulse. Laser sparks were made by focusing the second harmonic (532 nm) of a Nd:YAG laser (Quantel QY471) operating either as a Q-switched nanosecond laser (10-ns pulse duration) or as a pulsed mode-locked picosecond laser (30-ps pulse duration). Spark images were recorded perpendicular to the direction of the laser using a CCD camera system.

Spark energy measurements were made using a dual beam arrangement whereby a fraction (about 15%) of the laser energy was diverted to a reference detector and the remainder allowed to pass through the ignition chamber to a sample detector. The transmitted and reference pulse energies were measured using Laser Precision pyroelectric energy meters, which in turn were calibrated to a Scientech thermopile energy meter. The two energy levels were calibrated for zero spark energy by evacuating the reaction chamber to prevent breakdown. In the presence of gas, a zero difference between the transmitted and reference pulse energies was obtained in the absence of a visible spark. When breakdown occurred, the attenuation of the transmitted light relative to the reference light was attributed to absorption of energy by the spark. It has been previously determined that other losses such as radiance and scattering are negligible compared to the absorption [3].

Figure 1. Schematic of the experimental apparatus showing the principle of dual beam detection for measuring spark energies.
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However, especially in the case of ns laser breakdown, the pulse that emerges from the chamber can experience distortions in beam shape not present in the vacuum calibration measurements. Care was taken to collect all of the light by the energy meter to ensure accurate spark energy measurements. Spark energies were varied by attenuating the laser output using a polarizer and rotatable half-wave plate. The end-to-end uncertainty in the MIE measurements was conservatively estimated as ±25%. This uncertainty is due to both uncertainty in the spark energy measurement and the stochastic nature of the ignition process. This stochastic behavior has led some investigators [8] to report ignition probability for a given measured spark energy rather than a definite value of MIE.

The experiment was automated using a digital control unit and a 386SX computer. All tests were conducted at 1 atm and 298 K. Specified gas compositions of methane and air were metered into the combustion chamber using mass flow controllers and were allowed to circulate through the reaction chamber for a time sufficient to reach equilibrium (about 1 min). The control unit then closed the circulating valves, opened a bypass valve, and began a 1.6-s delay. A shutter was then triggered to allow a single laser pulse to enter the chamber to create the spark. The transmitted and reference energy measurements were read and digitized by an A/D card in the computer. Ignitions were detected by monitoring the pressure increase in the buffer chamber due to the rupturing of the Al foil membrane, in which case the control unit terminated the timing sequence and initiated an evacuation sequence.

**MIE MEASUREMENTS**

Minimum ignition energy measurements using ns and ps laser-induced sparks are presented in Fig. 2 for CH₄/air mixtures and are compared with previous electric

![Graph showing measured and calculated minimum ignition energies of CH₄-air mixtures at 1 atm. For comparison, also shown are results from electric spark ignition experiments by Lewis and von Elbe [1] and Ronney [12], laser spark ignition experiments by Kingdon and Weinberg [4], a numerical computation by Sloane and Ronney [2] and a simple hot gas model by Syage et al [3].](image)

Figure 2. Measured and calculated minimum ignition energies of CH₄-air mixtures at 1 atm. For comparison, also shown are results from electric spark ignition experiments by Lewis and von Elbe [1] and Ronney [12], laser spark ignition experiments by Kingdon and Weinberg [4], a numerical computation by Sloane and Ronney [2] and a simple hot gas model by Syage et al [3].
and laser discharge measurements and calculations. The most notable trends in Fig. 2 are summarized as follows:

1. The MIE curve for ps pulses lies at higher energy than for ns pulses, however, the difference decreases toward the lean and rich flammability limits.
2. The laser ignition results lie at higher energy than the electric discharge results, however, again the difference decreases toward the flammability limits.
3. The experimental results are bracketed by two model calculations. A simple hot gas model [3], based on the homogeneous heating of a minimum flame volume whose energy depends on the quenching distance for the mixture and a temperature specified through a chemical induction time, lies along the upper limits of the experimental values. The detailed computation by Sloane and Ronney [2] lies well below the lower limits.

We believe that the differences between our ns and ps laser results and between laser and electric spark results are larger than the uncertainties in the measurement of spark energy. In this context, it is worth noting that nanosecond pulses are comprised of a distribution of sharp spikes due to beating of the longitudinal and transverse modes of the laser cavity, whereas the ps pulses are much more uniform. Apparently, minute changes in the laser cavity can change these mode structures and alter the breakdown characteristics, though it is unclear at this point why this should affect the MIE significantly, since the time scale for hydrodynamic motion (i.e., of the blast wave created by the spark) is on the order of microseconds and the time scale for flame kernel evolution is on the order of milliseconds. We plan to examine these dependencies further in future work.

SPARK SIZE MEASUREMENTS

It was considered possible that the different minimum ignition energies by ps vs. ns sparks may be due to the dependence of ignition energy on spark kernel size [5]. For example, one might expect ignition to be less efficient if the spark size exceeds the critical energy deposition radius, which has been calculated as 0.03 cm for stoichiometric $\text{CH}_4$-air mixtures at 1 atm [2]. In order to characterize better the spatial extent of the ignition source, the laser-induced sparks were imaged using a slow-scan CCD camera system. A 50 mm camera lens with a doubler extension tube imaged the visible spark emission onto a 576 by 384 pixel, thermoelectrically cooled CCD chip. The spatial resolution in the plane of the spark was 0.0058 cm. The sparks were found to be intense compared to camera sensitivity and the lens aperture was stopped down to f/22 to enhance depth of field and minimize background contributions. The camera shutter was manually triggered with a two second exposure time to capture the image of a single laser-induced spark. We found sparks generated from nanosecond laser pulses to be much brighter than those generated from picosecond pulses for the same spark energy, which might have been expected considering the differences in breakdown mechanisms for the two laser sources (see Introduction). In order to avoid saturating the camera, a neutral density filter was used to attenuate the nanosecond spark emission by a factor of 100. We should note here that the use of visible emission as a diagnostic of spark size is not well established. Emission quenching near the edge of the spark or hot, non-emitting edges could lead to underestimates of size. Consequently, the current measurements are probably a lower bound on the spatial
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Figure 3. Contour plots with equal increments in intensity level (in arbitrary units) of the spark emission intensity profile for ns and ps laser breakdown. Spark energy is 5 mJ in both cases. For ps sparks the intensity scale has been multiplied by 100.

The extent of the energy deposition region. In future work, we will employ schlieren or interferometry to determine the region where significant heating occurs, and thus obtain an upper bound on the energy deposition region. Additionally, we plan to assess the size of the region where significant chemical reaction occurs, independent of spontaneous emission, using planar laser induced fluorescence to image hydroxyl radicals.

Figure 3 displays contour plots of sample images for picosecond and nanosecond pulse laser-induced sparks of 5 mJ nominal energy. Figure 3 suggests the picosecond pulse sparks are nearly spherical while the nanosecond sparks are elongated along the direction of the laser beam. In order to make a quantitative comparison of sparks generated from a variety of pulse energies and optical arrangements, we fit the spark image intensity profiles to a two-dimensional Gaussian with independently adjustable widths parallel to and perpendicular to the laser propagation axis. We found that the picosecond pulse sparks were well characterized by this functional form. The nanosecond pulse sparks were well characterized by a Gaussian in the direction transverse to the laser propagation, however, in the direction along the laser propagation, a shoulder appeared on the side closer to the laser source at spark energies above approximately 5 mJ. It should be noted that while fits to a single Gaussian were obtained to facilitate comparisons between picosecond and nanosecond pulse sparks, these sparks would be better characterized as a sum of Gaussians. Figure 4 summarizes results for both laser sources and for a range of spark energies in terms of the half width at half maximum (HWHM) of the sparks parallel and perpendicular to the laser beam. The width of the sparks transverse to the laser is essentially independent of the pulse energy for both laser sources. There is a slightly smaller transverse width for the ps sparks (HWHM=0.018 cm) compared to the ns sparks (HWHM=0.022 cm). A weak trend toward increasing width along the laser beam with increasing laser energy is observed for the ns sparks, but to a good approximation the spark size is independent of the pulse energy.
DISCUSSION

Using detailed models of the chemistry and transport processes occurring in a spherical flame kernel, Sloane and Ronney [2] predicted that the critical energy deposition radius \( r^* \) for stoichiometric \( \text{CH}_4 \)-air mixtures is 0.03 cm. For an energy deposition radius \( r < r^* \), the MIE is essentially independent of \( r \), whereas for \( r > 0.03 \) cm, the MIE is proportional to \( r^3 \). Our measurements of spark kernel size indicate that for ps sparks \( r < r^* \), and thus \( r \) is small enough that the MIE should be independent of \( r \), and for ns sparks \( r \) is only slightly larger than \( r^* \). Consequently, we would expect the MIE to be close to the 0.1 mJ predicted by Sloane and Ronney. However, the MIE measurements do not support this conclusion; for the ps laser the MIE is about 25 times larger than this value and for the ns laser the MIE is about 10 times this value. For ps sparks, the fact that the MIE is about 25 times larger than the computationally predicted value would suggest \( r/r^* = 25^{1/3} = 3 \), thus \( r = 0.09 \) cm. While it seems unlikely based on Figs. 3 and 4 that \( r \) was as large as 0.09 cm, the data shown in these figures are lower bounds on \( r \). Thus it is possible that for stoichiometric mixtures, \( r \) is too large to obtain the MIE predicted by Sloane and Ronney. The difficulty of obtaining sufficiently small ignition kernels and the effect of too-large kernels is discussed further elsewhere [5].

For mixtures that are leaner or richer than stoichiometric, \( r^* \) increases substantially from its value for stoichiometric mixtures. Evidence of this is readily seen in the effect of mixture strength on the optimal electric spark gap setting [1]. Furthermore, theoretical studies of ignition [9] show that \( r^* \) scales with the flame thickness, which is inversely proportional to burning velocity which in turn decreases rapidly for mixtures lean or rich of stoichiometric [10]. In the current experiments, for compositions between 10.4% and 6.1% \( \text{CH}_4 \), measured values of MIE change by less than a factor of two. On the other hand, \( S_b \) decreases from 40 to 15 cm/sec for these mixtures [11], and thus for these mixtures the ratio of MIEs, which would be proportional to the ratio of the ignition kernel volumes, should be roughly \((40/15)^3 = 19\). This suggests that \( r \) is in fact too large to obtain the true MIE for these mixtures. Additionally, for very lean or very rich mixtures, the measured values of MIE start to increase substantially, which would be consistent

with the large \( r \) investigated here. Thus, the effect of \( r \) is

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SUMMARY

The critical energy deposition radius \( r^* \) was revisited, and the total ignition energy was shown to consist of two parts. For ns sparks, these parts were

Figure 4. Compilation of spark width and length relative to the laser propagation direction as a function of spark energy for ns and ps laser sources.
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with the notion that for these mixtures, which have low burning velocities and thus large $r^*$, $r < r^*$. Furthermore, as shown in Fig. 2, MIE data from three different investigators [1, 4, 12] that employed different types of ignition sources having different $r$, all converge to nearly the same values for lean mixtures with large $r^*$. Thus, one explanation for the MIE behavior found in this study is related to the effect of the ignition kernel size on MIE, however, this does not explain the differences between the ns and ps laser results.

Another potential mechanism that would render the measured MIE larger than calculated values is the effect of gasdynamic shock losses. The electric or laser spark energy deposition process creates a kernel of hot, high pressure gas that produces a quasi-spherical shock wave as it expands. It is possible that energy can be convected by gasdynamic processes to radii larger than $r^*$, which would increase the MIE over that if the shock wave dissipated at a radii less than $r^*$ and all the energy were retained within this region. While this loss mechanism has been proposed by others [1, 4], quantitative estimates were not provided. According to the classical Taylor blast wave model [13] of the response of a gas with specific heat ratio 1.4 to an instantaneous deposition of energy in a very small volume, the energy left behind in the form of internal energy of the gas is close to 1/3 of the initial energy deposition. Thus about 2/3 of the total laser spark energy could be lost to shock losses if the energy deposition volume were very small. This is only an upper bound on the losses, however, because in the experiments the energy deposition radius was not infinitesimal but rather was comparable to $r^*$ for near-stoichiometric mixtures. Also, because the extremely high temperatures generated by the laser spark will lead to dissociation of the gas, some energy will be left behind in the form of dissociated molecules that will liberate thermal energy as they recombine. Thus, it would seem that no more than perhaps 50% of the laser spark energy could be lost to shock losses. Further evidence for this assertion can be found in the work by Frendi and Sibulkin [13], who employed an ignition model employing detailed gasdynamics but simplified chemistry and transport properties and found that the constant-pressure MIE could be at least a factor of two lower than that computed when shock-wave effects were included. Thus, while the shock losses may be substantial, they cannot by itself account for the discrepancies between different ignition sources or the discrepancies between the experiments and numerical calculations assuming isobaric heat input [2].

While the asymmetries of the laser spark might be expected to affect the MIE, it is difficult to attribute the discrepancies between models and experiments observed in this work to this cause. Note that the larger, more asymmetric ns sparks had MIEs lower than the smaller, more symmetric ps sparks. Moreover, the ignition kernels produced by electric spark ignition sources are at least as asymmetric as the ns laser sparks employed here and the heat losses to spark electrodes due to the large cathode and/or anode voltage drops should render the electric sparks less efficient ignition sources.

SUMMARY AND CONCLUSIONS

The classical minimum ignition energy experiments of Lewis and von Elbe are revisited using a laser ignition source that enables accurate characterization of both the total energy deposition and the spatial extent of the deposition. Experiments show the expected trends in minimum ignition energy and are quantitatively consistent with other measurements for CH4-air mixtures far from stoichiometric. For near-stoichiometric mixtures the measured values are higher than expected. These results may be a consequence of the size of the energy deposition region with
some possible influence of gasdynamic shock losses. In future work efforts will be made to reduce the size of the deposition region and determine an upper bound on the size of the region where energy is deposited. Furthermore, the ns and ps laser sparks will be characterized further in an attempt to determine the reason for the discrepancies between the MIEs measured using these two ignition sources.

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