Laser-induced ignition of methane and biogas near the lean flammability limit

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Abstract

Interest in fuel flexible combustion systems necessitate characterization of various fuels with respect to their combustion properties, such as ignition energy requirements. In this work we study laser ignition of methane and biogas near their lean flammability limits. A high energy Nd: YAG laser at 532 nm is used to induce breakdown and ignition of fuel/air mixtures. Plasma formation, flame initiation, quenching, and successful flame propagation are captured using high speed Schlieren imaging and laser interferometry. Minimum pulse and minimum ignition energies are determined for a range of equivalence ratios, also permitting the determination of flammability limits. These measurements offer insight into differences in bio and methane ignition such as the higher energy requirements for biogas. The results and discussions advance our understanding of ignition requirements for various fuel/air systems.

1 Introduction

Laser ignition is of interest as a possible alternative ignition source for combustion systems. Multiple benefits are associated with laser ignition such as optimized ignition placement, reduced maintenance in applications with long operational times, and extending lean combustion limits. Strides towards cleaner, sustainable energy production are also being taken in the area of alternative fuels, particularly the design and operation of combustion systems that incorporate fuel flexibility. This makes it possible to use biogas in systems designed to use traditional fuels. Biogas is typically a combination of 15-40% carbon dioxide and 60-80% methane. The addition of CO_2 leads to different performance characteristics. To ensure proper performance, ignition systems must be calibrated for variability in combustion mixtures.

The first step in characterizing ignition performance of new fuels for laser ignition is to determine ignition thresholds. Minimum ignition energy (MIE) is one of the most important parameters in the ignition characterization of fuels. In a similar light, minimum pulse energy (MPE) is important from a technical standpoint. Lower MPE means a smaller laser can be used for ignition, lowering costs. Previous studies have shown breakdown and ignition thresholds depend on a number of factors including laser characteristics, pressure, temperature, and focusing optics. It is now well known that MIE decreases with prevailing pressure and temperature. Although these trends may be consistent and independent of fuel, the variability of MIE across other parameters such as equivalence ratio cannot be predetermined.

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Previous work in laser ignition is extensively reviewed by Ronney [1] and Bradley et al. [2] giving fundamental descriptions of plasma breakdown. Further, Morsy [3] reviewed application aspects in the context of current technological capabilities. Research activity in this area has focused largely on combustion of methane. Experiments by Ma et al. [4] using a single cylinder test engine showed laser ignitions feasibility and performance improvements. Phuoc et al. [5] characterized methane MIE measurements finding ignition energies of 3 to 4 mJ near stoichiometric conditions and as high as 40 mJ at the lean limit. The dependence of MIE on parameters, such as focal length and pressure, was investigated by Kopecek et al. [6]. Srivastava et al. [7] have recently added investigations of the structure and flame propagation of compressed natural gas ignition. Despite the wealth of literature pertaining to methane ignition, few studies have compared laser ignition of methane with biogas. Forsich et al. [8] made the first comparison for fuel rich to fuel lean biogas at pressures up to 3 MPa. The study focused on water production near the spark location and flame emissions, leaving out a detailed investigation into ignition energy. Recently, Biet et al. [9] investigated ignition energies for methane and biogas for extreme fuel lean conditions. Comparisons were made with electrode spark ignition near the lean flammability limit.

In this study we further investigate the differences between laser ignition of methane and biogas. We first explore the effect of key parameters on breakdown threshold including pressure and focal length. Focus is then turned to reactive mixtures where comparisons are made of two ignition energies, MIE and MPE. Finally, we investigate flame formation near the lean flammability limit using two diagnostic techniques. Most of the studies previously mentioned used Schlieren imaging to visualize plasma and flame development. Here we use laser interferometry [10] to further analyze the ignition process.

2 Experimental approach

Experiments are carried out in a cylindrical stainless steel chamber with optical access on six sides. The chamber is 15.24 cm in diameter and 25.4 cm long. Methane, CO_2 , oxygen, and nitrogen are delivered through a central manifold system. To fill the chamber, it is first evacuated of all contents. Mixture preparation is done through the partial pressure technique. The gases are allowed to mix and settle before the test. The biogas in this study is a mixture of 60% methane and 40% CO_2 .

Mixtures are ignited using a Spectra-Physics 10 Hz Nd:YAG laser at 532 nm with a pulse duration of 10 ns. Laser energy is measured in two locations using power meters (Ophir PE-25). One power meter determines the incident energy from a portion of the beam deflected by a beam splitter. The other power meter is placed behind the chamber to determine the residual energy after breakdown or ignition. The laser energies recorded by the power meters are corrected to the true value inside the chamber by taking into account losses through the sapphire windows. To ensure the accuracy of the energy readings, the chamber was vacuumed out before each test and the laser was pulsed to guarantee nearly 100% transmission. Uncertainties in the readings by the power meters are determined to be less than 3%.

Schlieren images are recorded with a high speed camera (Photron SA-4) operated at 20,000 frames per second. The Schlieren setup is a LED light source coupled with an optical fiber and collimated between two 50 cm focal length mirrors. All data acquisition and process control are done using a computer program. The camera is triggered by a digital delay generator (SRS DG-645) that is timed with the laser pulse. Laser interferometry is performed using a He:Ne laser with optics configured in a Mach-Zehnder interferometer arrangement. Images are recorded with the same high speed camera described previously.

3 Laser breakdown, energy absorption and focal length dependance

The homogeneous gaseous mixture is normally transparent to the laser light at 532 nm. This is shown in Figure 1 where incident energy and energy transmitted through the focal volume are plotted. At



Figure 1: Transmitted verse incident energy at 0.45 atm and 1 atm and 298 K.



Figure 2: Breakdown threshold for 150, 200, 250, and 300 mm focal lengths in air at 1 atm, 298 K.

energy levels below the breakdown threshold the trend is linear along the dashed line showing incident energy equal to transmitted energy. The data points depart from the linear trend above the breakdown threshold where a higher percentage of energy is absorbed. From this, energy absorption clearly relies on the formation of plasma. Larger absorption percentages occur for more highly ionized plasmas. Plasma formation is possible through multi-photon, electron cascade, and penning ionization processes. Penning ionization [11, 12] is observed in two or more gases with differences in ionization potentials. Also seen in Figure 1 is the pressure dependance on breakdown threshold. At higher pressures, a lower incident energy is required to create plasma due to more collisions during electron cascade since there are a larger concentration of molecules in the focal volume.

Gas dielectric breakdown and plasma formation is related to the focal volume formed by the focusing optics. The focal volume is strongly dependent on the focal length of the lens used. An electric field strength or power density is needed to effect the breakdown of the non-conducting gas. As the focal volume decreases, the power density will increase, so that breakdown is able to occur with a lower incident energy. Figure 2 shows the effect of focal length on the breakdown of air. It is observed that the incident laser energy required to create breakdown increases with the focal length.

4 Ignition energy and flame formation

In this study we compare two measurements of ignition energy of biogas and methane mixtures, MIE and MPE, from lean to stoichiometric conditions. Figure 3 shows the dependence of MPE on equivalence ratio. It is observed that at all equivalence ratios investigated, a higher MPE is needed to ignite biogas compared with methane. CO_2 acts as an additional diluent in biogas leading to more energy required to initiate combustion. Near stoichiometric conditions, the MPE was found to be around 16.5 mJ for methane and 24 mJ for biogas. The difference in MPE between the two fuels stays consistent at 6-10 mJ across the range of equivalence ratios. Figure 4 shows the MIE for methane and biogas. Again, biogas requires a higher energy for ignition. The MIE for near stoichiometric conditions was around 2.25 mJ for methane and 4 mJ for biogas. As apposed to MPE, the difference in MIE for the two fuels is nearly double for every equivalence ratio. This difference is small where the ignition energies are low, but leads to very high energy absorption for biogas compared with methane at extremely lean conditions.

Ignition can be a sporadic event close to lean limits. Mixtures ignited under the same conditions near the lean limit show an increase in the variability in the energy at which they ignite and flames have a



Figure 3: MPE at 1 atm, 298 K.

Figure 4: MIE at 1 atm, 298 K.



Figure 5: Schlieren images of quenched flame and ignition of biogas mixture, $\phi = 0.65$, with incident energies 27.5 mJ and 28 mJ, respectively.

greater probability of quenching. Flame quenching is an area of concern for combustion systems since it can lead to misfires in internal combustion engines and flame blowout in gas turbines. This necessitates further investigation into the mechanisms controlling ignition in this regime.

Figure 5 shows a quenched flame and successful combustion for a biogas mixture at $\phi = 0.65$. The early stages of breakdown and flame kernel development look similar. In the second image at 1.75 ms, it is observed that density gradients in the front lobe begin to grow weaker for the quenched flame. The successful event eventually leads to complete combustion of the fuel in the chamber, even though only 0.5 mJ of incident energy separate the two cases. With Schlieren imaging, density gradients are visualized as changes in light intensity making it difficult to see subtle differences between two events. To further investigate the ignition process of these fuels, we employ interferometry. Here, deflections in the interference fringes show the magnitude of the change of refractive index in a medium due to an alteration in the density. These density changes can arise from a number of factors including temperature, pressure, species concentration, and electron density of a plasma. For a complicated phenomenon like laser ignition, identification of the source of the refractive index change can be difficult since many of these factors are coupled at certain times throughout the combustion process.

Figure 6 shows interferometric images of laser induced breakdown in air and methane ignition and



Figure 6: Interferometer images of breakdown in air, ignition of methane ($\phi = 0.6$), and ignition of biogas ($\phi = 0.6$), at an incident energy of 21.5 mJ and T = 298 K, p = 1 atm.

biogas ignition at an equivalence ratio of 0.6. All of these occur around the same energy level of 21.5 mJ. The first image at 5 μs shows plasma formation in the center of the image surrounded by a shock wave. Analyzing the fringe deflections, the plasma and shock formed in the methane and biogas mixtures are nearly identical. For breakdown in air, the shock shows slightly higher refractive index changes while the refractive index change at the center of the plasma is slightly lower than the methane and biogas mixtures. One explanation for this may be that the methane and biogas mixtures have a higher electron density in the plasma formed due to the fact that they have a lower ionization potential than air by itself. At 0.13 ms the fringe deflections for methane and biogas are again nearly identical. Fringe deflections for the plasma created in air begin to diverge more heavily from the reactive cases as flame kernel formation begins. Finally, at 1.30 ms the fringe deflection in the air breakdown case is very low as the free electrons have mostly recombined with the ionized molecules. By this time the methane and biogas flame kernels have diverged to a point where they show very different changes in refractive index with the methane flame kernel appearing much stronger. For these ignition events, the methane flame eventually leads to complete consumption of the fuel in the chamber while the biogas flame quenches, as indicated by the images at 5 ms. With this analysis we can more clearly see where these two ignition events differ. It is not the plasma processes early on that seem to hinder the ignition of the biogas. Instead, it is when the transition to a flame kernel begins as the plasma recombines and chemical reactions dominate the development that the addition of CO₂ in the biogas hinders flame growth.

5 Concluding remarks

Ignition energies and flame propagation of methane and biogas mixtures at a laser wavelength of 532 nm, initial chamber pressure of 1 atm and temperature of 298 K have been investigated. MIE and MPE are found to be higher for biogas. MIEs for biogas are nearly twice those of methane at similar

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equivalence ratios. This leads to high energy absorption in biogas compared to methane at extreme lean conditions where MIE is large. Pressure and focal length effects have also been studied for breakdown energy in air. As shown in literature, the breakdown energy threshold decreases as pressure increases. The breakdown energy threshold increased at longer focal lengths.

Flame development for quenched and ignited methane flames was analyzed using Schlieren imaging. Although the quenched and ignited cases started the same, they deviated from each other at later times. Detailed investigations of the ignition process are difficult with these images highlighting a drawback of Schlieren imaging. Laser interferometry was used to investigate air breakdown, methane ignition, and biogas ignition at similar energy levels. The usefulness of interferometry as a diagnostic tool for ignition studies was highlighted by obtaining subtle differences between the ignition and breakdown processes that would be difficult to obtain otherwise. This work contributes to the development of fuel flexible combustion systems by characterizing differences in the laser ignition of bio- and natural gas.

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