Gas dynamics of laser ignition in butane-air combustible mixtures

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Abstract. The advantages of laser ignition compared to traditional electric spark are in optical breakdown threshold, being inversely proportional to pressure up to hundreds of bars; and in the possibility to reduce harmful emissions (NOₓ, CH, etc.) at fuel lean mixtures ignition. Laser-induced combustion waves are known to appear faster and are more stable than when induced by electric spark, so can also be used for ramjets, rocket thrusters, and gas turbines. Laser ignition occurs as a result of gas breakdown, formation of plasma and shock waves leading to the combustion core formation. For butane-air mixtures (p~1÷3 bar) of different compositions (φ~0.8÷1.4), the dynamics of the combustion core and shock wave propagation induced by laser ignition (1064 nm, 12 ns) with 200 mJ pulses was studied. Using schlieren photography, propagation velocities of shock wave front and combustion core boundary were measured within 0.2÷4 µs after exposure.

1. Introduction

Ignition is a complex phenomenon that strongly influences the subsequent combustion. At early stages it strongly influences the formation of pollutants, flame propagation and collapse, and detonation. Lasers have been widely discussed as a potentially superior ignition source for industrial appliances. Advantages of laser ignition in comparison with traditional electric spark are: faster combustion core development [1] and absence of electrode or other confinements spoiling it. The performance of engines with laser spark plugs [2] has been studied for several years already [3–5]. Mainly methane has been investigated as a modern clean fuel, and hydrogen as a perspective one. However, very few works have shown the actual dynamics of laser-induced combustion. Among those who did, most were interested in micro- and millisecond range after impact [6–8]. While early sub-microsecond stages were considered in [9] only.

The blast wave theory characterizes the temporal evolution of a shock front from a point-like rapid deposition of energy. Given the energy, the theory predicts the time dependence of the shock front position. And vice versa, from the temporal evolution of the shock front, the energy used to generate the blast can be estimated. Original Sedov-Taylor intense explosion theory [10] is not supposed to be applied to the cases when Mach number quickly reduces to near unity, which is the case for laser-induced shock waves.

Our study focuses on the experimental study of the early stages of laser-induced combustion, which is the time interval between several hundred nanoseconds (breakdown and energy transfer from laser to plasma) to several microseconds (generation and propagation of a shock wave). Another feature is that butane based mixtures are studied as those are similar to liquefied petroleum gas (LPG) – a fuel
preferable for automotive engines since much more heat can be stored in a unit volume, and it could be liquefied at 20–30 bar pressures – an order of magnitude lower than methane and hydrogen [11].

2. Experimental layout
The experimental layout is presented in figure 1, it was described in details in [12]. We have used butane from commercial gas burner cartridges (propane – 6%, butane – 28%, isobutane – 60%, impurities – 6%) to make mixtures of different equivalence ratios (φ~0.8÷1.4, and φ=0 for reference) at pressures (p~1÷3 bar). A combustible fuel-air mixture was prepared in an evacuated combustion chamber by letting in fuel and then purging compressed air. After ignition, pressure in the chamber raised rapidly, when it reached ~5 bar, the blow-off valve was automatically opened, and exhaust gases from the combustion chamber were drained to an elastic tank. The fundamental frequency radiation (1064 nm) of a nanosecond (12 ns) Nd:YAG laser (Solar LS LQ929) was focused by F=150 mm double convex lens to induce a spark. Ignition pulse energy was about the same for each shot (200 mJ).

Schlieren photography was used to visualise the shock wave formation and flame kernel development following the laser-induced breakdown and plasma formation since it considerably improves the contrast [13]. An ICCD camera (Nanoscan Nanogate-2) was used to capture images at 10 ns exposure gate. Probing was performed by 473 nm DPSS-laser (Lasever LSR173NL200) radiation; a 20 nm bandwidth interference filter was mounted before the sensor to cut-off plasma broadband radiation. We observed considerable carbonisation of optical windows after combustion. This did not affect igniting laser radiation transmission due to soot ablation by pre-pulse, but strongly affected probing. At 3 bar, dense dew was observed at these windows.

3. Results and discussion
Evaluation of shock wave source energy was performed following [14]: 
\[ E_s = R^5 \rho / \psi^5 t^2 \], where \( R \) is shock wave radius; \( t \) is time after impact; \( \rho \) is undisturbed gas density; \( \psi \) depends on gas adiabatic index, and in our case was equal to 1.01. A series of reference images was taken for each pressure and laser pulse energy for pure air without combustible gas. We have measured radial and axial dimensions of shockwave and combustion core (plasma core without fuel), ignition centre position and number of ignition spots along the optical axis (figure 2).

Figure 1. Experimental setup (1 – combustion chamber, 2 – Nd:YAG laser, 3 – probing laser , 4 – ICCD camera, 5 – gas bottle, 6 – vacuum pump, 7 – valves, 8 – air compressor, 9 – spectrometer, 10 – optical fiber, 11 – membrane vacuum gauge, 12 – acquisition unit, 13 – temperature controller, 14 – high pressure sensor, 15 – oscilloscope, 16 – energy meter, 17 – beam splitter, 18 – lens, 19 – telescope, 20 – imaging objective, 21 – edge, 22 – interference filter).
The mechanisms leading to the flame core asymmetric growth are discussed in [8, 9, 15]. Need for two- or multi-centre consideration of this phenomenon was stated in [16]. Initially, shock wave has an egg shape with the major axis orientated along the laser beam due to asymmetric laser energy deposition, and becomes spherical later on.

Noteworthy, for laser-induced spark, the plasma core radius, measured for 1 μs delays, was almost the same for different equivalence ratios. This indicates that the influence of chemical heat release on the initial size of the nucleus is negligible. Similarly, it was found in [17] that φ has a minor effect on the ellipsoidal core size for methane/air mixtures at 1 bar. But in case of electric spark, at breakdown energy of 0.8 mJ [8], equivalence ratio had an appreciable effect on the initial size of the flame kernel [18]. That could be explained by greater sensitivity of ignition development just above its threshold and in presence of confinement.

Initial shock wave velocity is higher than that of the flame front. When the shock wave detaches from the hot air core, both phenomena can be studied independently. Combustion core was following the shockwave front until ~1 μs, then collapsed slightly and stayed almost the same size until the end of observed delays range (4 μs). There was no qualitative difference observed to laser breakdown in pure air. Quantitative difference was reported in [6]: about 0.5 ms after impact, shock wave propagation velocity became greater for combustible mixtures than for pure air; that suggests the evidence of chemical energy release.

Source energy was estimated following the intense explosion theory [10, 14] for radial component of plasma core evolution for each schlierengraph in delays range of 200 ÷ 900 ns, and then averaged for each experimental series (figure 3). The results presented in table 1 show significant difference in estimated blast wave source energy between reference experiments in pure air and combustible mixture. Standard deviations presented in table 1 are relatively big, but it should be mentioned, that such highly indirect measurement accuracy estimate is at least 40%. Laser pulse energy in our case was much greater than theoretically necessary, an order of magnitude greater than the minimum ignition energy [19, 20]. Conventional electric spark energy also usually exceeds this value to prevent misfires.

In [8] it has been suggested that no energy should be released from combustible gas mixture at this timescale. Laser pulse energies used in that work were 5 ÷ 20 times lower. However, the discrepancy in plasma core radius and theoretical predictions was ~30%, that corresponds to 5 times more powerful source than just laser spark, so no difference suggestion does not seem to be well proven in such case.
Table 1. Blast source energy estimate (mean±SD, n=7–10 for each series).

|       | p= 1 bar         | p= 2 bar         | p= 3 bar         |
|-------|------------------|------------------|------------------|
| φ=0   | 151.5±56.9 mJ    | 97.5±29.8 mJ     | 97.3±44.3 mJ     |
| φ=1   | 306.6±65.8 mJ    | 202.7±66.3 mJ    | 228.2±133.2 mJ   |

Figure 3. Comparison of plasma core radial diameter for φ=0 (1, 2, 3) and φ=1 (4, 5, 6).
1, 4 – 1 bar; 2, 5 – 2 bar; 3, 6 – 3 bar.
Inset shows a characteristic schlieren graph (φ=1, 2 bar, 1 μs)

Figure 4. Multiple breakdown regions at φ=0.9, 3 bar
a – 0.2 μs; b – 0.4 μs; c – 1 μs

Our results show that pulse energy could be sufficient to start detonation and so release chemical energy. Plasma core speed in this case matched well referenced data for butane detonation (~4 km/s).
Analysis of data on breakdown centre position instability has not revealed any clear dependencies. However, for 2 and 3 bar pressures dispersion was always 1.5÷2 times lower than at 1 bar. We have observed 2 breakdown regions with ~10% probability at 2 bar, and 2 or more such regions in ~30% cases at 3 bar (figure 4). Blast source energies presented in table 1 in case of multiple breakdowns were summarised.

Shock wave centroids were always displaced up the laser beam compared to plasma core centres. The implication that the shock-wave originates closer to the leading than to the trailing edge of the plasma ellipsoid is explained by laser energy absorption resulting in radiation intensity exponential decay. Similarly, the centre of the spherical rarefaction wave is always upstream of that of the plasma core [6].

4. Conclusions

The dynamics of combustion core at laser ignition of butane-based mixture specifically, has been studied for the first time in 0.2–4 μs delays range after impact. It has been shown that at comparatively high laser pulse energies detonation can start and contribute to the blast wave source. Blast source energy increase observed at the impact on stoichiometric mixture could evidence this. The influence of fuel mixture equivalence ratio on the flame behaviour and extinction (initial size of the flame core) was found to be minor in contrast to the electric spark.

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