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Ignition of hydrogen–air mixtures using pulsed nanosecond dielectric barrier plasma discharges in plane-to-plane geometry

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ABSTRACT

Ignition of preheated (400–500 K) H_2–air mixtures at low pressures (80–100 torr) excited by pulsed nanosecond dielectric barrier discharges is investigated through experiments and simulations. Time resolved absolute OH concentration and temperature data are obtained using Laser Induced Fluorescence (LIF) technique. Ignition is achieved in the decaying plasma after a burst of discharge pulses (repetition rate 10–40 kHz), with the time delay inferred from sudden rise in OH^* emission. One-dimensional simulations are performed to obtain information about the plasma generated radicals and heat release across the discharge gap. A plasma fluid formulation is used with ions and neutral species at gas temperature, and electrons in non-equilibrium. An accurate reduced chemistry mechanism is developed through sensitivity analysis to expedite the plasma simulations. The model predictions show excellent agreement with experimental measurements, validating the numerical framework and chemistry data. The input pulse energy and ignition characteristics are found to be highly sensitive to uncertainties in dielectric properties. Ignition delay exhibits a threshold-like dependence on input plasma energy, and increases steeply as the number of pulses in the burst is reduced. The nanosecond plasma assisted ignition is achieved through a two-step process. Firstly, the burst of discharge pulses produce a large pool of radicals and provide an average temperature rise of ~1–2 K/pulse. In the next step, if the temperature exceeds a threshold value of ~700 K, significant heat release from partial fuel oxidation is triggered. The process becomes self-sustaining and the temperature continues to rise even after the plasma source is switched off, accelerating the conventional H_2–O_2 chain branching pathways and leading to ignition. We provide conclusive evidence of large volume ignition with nanosecond plasma as opposed to thermal ignition at a hot-spot. Ignition is first observed at the center of the discharge gap, but the kernel expands rapidly to the entire volume, except near walls where heat losses keep the temperature low. It is demonstrated that the ignition occurs independently at different locations due to local plasma chemistry effects and heat transport does not play a significant role.

1. Introduction

Non-equilibrium plasma discharges (NPD) have shown great promise in thermal and kinetic enhancement of ignition and combustion at a wide range of pressures, flow velocities and equivalence ratios [1–3]. Experiments have demonstrated successful ignition and flame stabilization with plasma assistance at the low pressures and small flow residence times encountered in high-speed propulsion systems [4–6], as well as at the high pressures (1 to several bars) relevant to gas turbine and IC engine applications [7–9]. A variety of NPD systems such as pulsed coronas [10], microwave discharges [11], magnetic gliding arcs [12], and pulsed nanosecond volume discharges [1–4] have been studied for potential combustion applications. High-voltage, nanosecond duration pulses applied at kHz repetition rates create volumetric plasma with E/N (ratio of electric field magnitude to number density) in the range of 100–1000 Td (1 Td = 10^{-17} V cm^2) [3]. The discharge develops in the form of fast ionization waves with a large fraction of input energy consumed in molecular dissociation and excitation of internal energy modes. The pulse duration (10–100 ns), is much shorter than the characteristic timescales for development of ionization instabilities. These features allow nanosecond discharges to generate a large number of active radical species at much higher pressures and power loadings, and lower power budgets as compared to other plasma discharge systems [3].

The nanosecond plasma can be a source of heat and reactive chemical species. Starikovskii and co-workers [13–15] performed shock tube experiments in which a fuel–air mixture was preheated by a shock wave above the ignition temperature. Radicals

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generated by a single nanosecond pulse discharge triggered chain branching processes leading to reduction in ignition delay by 1–2 orders of magnitude for a variety of fuels. The additional heating produced by the discharge pulse played only a minor role.

Over the past few years, the flow reactor experiments described in Refs. [16–20] have focused on understanding ignition of fuel–air mixtures excited by pulsed nanosecond plane-to-plane dielectric barrier discharges (NS DBDs) under mildly preheated conditions (100–200 °C). Intensified charge coupled device (ICCD) images demonstrate that pulsed NSDBDs are uniform and devoid of any filaments in air [16] and preheated H_2–air [17–20] in 40–100 torr pressure range. Discharge uniformity is necessary, if non-equilibrium chemistry is to be separated from thermal effects occurring in filaments and hot-spots. In addition, a diffuse volume filling plasma facilitates optical diagnostics and experimental reproducibility. In this configuration, ignition delay [17] and OH concentration [18] were measured in mildly preheated (100–200 °C) H_2–air mixtures subjected to nanosecond voltage waveforms. The number of pulses leading to ignition was found to be a weak function of mixture equivalence ratio, but showed a non-linear dependence on pulsing frequency. At a given temperature and pressure, there existed an optimum repetition rate at which the number of pulses needed for ignition reached a minimum. Reduction in coupled pulsed energy at high repetition rates, due to increase in residual electron density, has been suggested as a possible cause for this behavior. Recent pulse energy measurements in air [16] did not show significant reduction in coupled energy at high repetition rates, suggesting a more comprehensive kinetic analysis is necessary to explain this phenomenon.

In Yin et al. [17,18], sustained application of voltage pulses at a high repetition rate "masked" the sensitivity of ignition characteristics to the details of plasma kinetics. In order to circumvent this behavior, experiments were recently conducted in a decaying plasma after a burst of discharge pulses [19,20]. The ignition characteristics were found to be highly sensitive to temperature and radical concentration at the end of a pulse burst. Ignition delay increased steeply as the number of pulses in the discharge burst was reduced. Kinetic model calculations showed that rise in temperature and H atom concentration beyond a critical threshold triggered chain reaction pathways leading to ignition. The combination of thermal and kinetic effects resulted in a reduction of threshold ignition temperature by ~200 K as compared to thermal ignition. It should be noted that the species and temperature measurements in the plasma flow reactor experiments were conducted in the central region of the discharge volume. The heating and species production rates near the plasma boundary (sheath) layers, however, are uncertain.

High-fidelity numerical models coupled with detailed chemistry mechanisms can complement experiments in study of thermal and non-thermal aspects of plasma assisted ignition and combustion. The wide disparity in timescales offer enormous computational challenges, and it is difficult to resolve important physical phenomena without compromising the accuracy of the results. A self-consistent multi-scale numerical framework for studying the spatio-temporal evolution of nanosecond plasma over multiple voltage pulses was established in [21]. One-dimensional simulations of pulsed NS DBDs in air were conducted in a plane-to-plane geometry in the pressure and pulsing frequency range of 40–100 torr and 1–10^5 Hz respectively. The model predictions were validated against experimental measurements [16] and results from a quasi-1D analytical model [22] of nanosecond discharges. The input electrical energy remained fairly constant on a per molecule basis from pulse to pulse. Repetitive pulsing generated atomic oxygen in the discharge volume via electron impact dissociation during voltage pulses, and through quenching of excited nitrogen molecules in the afterglow. Ion Joule heating during each voltage pulse caused a rapid rise of temperature (30–40 K) in the cathode sheath layers. Wall heat loss in the time interval between discharge pulses, however, prevented overheating near the boundaries. The volumetric heat release from quenching of excited species resulted in a "hat shaped" temperature profile after multiple discharge pulses.

In the present work, we perform comprehensive experimental and numerical investigations of pulsed nanosecond plasma assisted H_2–air (mildly preheated) ignition in a plane-to-plane geometry. Time resolved OH concentration and temperature are obtained from LIF measurements in the center of the discharge volume. Ignition is achieved in the decaying plasma after a burst of discharge pulses. One-dimensional simulations are performed to obtain information of the plasma generated radicals and heat release across the discharge gap. The uniform and diffuse nature of the preheated H_2–air plasma [16–20] justifies the 1D assumption. The accuracy of the model is assessed by comparing against OH concentration, temperature and ignition delay measurements after a burst of discharge pulses [19,20]. An accurate reduced chemistry mechanism is developed through sensitivity analysis to expedite the plasma ignition simulations. The sensitivity of input pulse energy and ignition characteristics to residual electron density and dielectric properties is analyzed. Electric field transients during each nanosecond pulse, and spatial evolution of gas temperature, excited species and radicals until ignition (ms timescales) are studied in detail. A special focus of this work is the question whether nanosecond pulsed plasma can produce volumetric ignition. The growth rate of the ignition kernel is studied in detail, investigating the roles played by heat transport and local radical chemistry.

2. Experimental setup

A schematic of the experimental setup used in the present work is shown in Fig. 1. It is similar to the one used in previous studies and described in detail in [18–20]. Briefly, a rectangular cross-section quartz channel (280 mm length, 22 mm breadth, 10 mm height, wall thickness 1.75 mm) is used as the discharge cell. Two plane quartz windows are fused to the ends of the channel, providing optical access in the axial direction. The entire assembly is heated in a tube furnace to improve plasma stability. The fuel and air flows are premixed before the cell inlet. The flow rates through the reactor are controlled by mass flow controllers. Two rectangular copper plate electrodes (14 mm width and 60 mm length) are placed on the top and bottom of the quartz channel. In order to reduce air gaps and prevent corona discharge outside the discharge cell, a 1/16 inch. thick high-temperature dielectric sheet (perfluroelastomer – Kalrez, DuPont) is placed between each electrode and the channel wall.

The electrodes are connected either to a Chemical Physics Technologies (CPT) or to an FID GmbH high voltage pulsed power supply. The CPT pulser is capable of producing 25 kV peak voltage, 25 ns duration pulses at repetition rates up to 50 kHz. The FID pulser can produce 30 kV, 5 ns pulses up to 100 kHz repetition rates. When CPT pulser is connected to the electrodes, the discharge cell is irradiated by a UV emission source (Hg–Ar lamp) to produce breakdown by the first discharge pulse. No pre-ionization is necessary when FID pulser is in operation. Ignition is detected by monitoring OH^* emission through the cell side wall with the help of a narrow bandpass filter (centered at 310 ± 2 nm, bandpass 10 ± 2 nm FWHM), a photomultiplier (PMT) and a digital oscilloscope.

The OH LIF apparatus is similar to one used in [18] and consists of a Nd:YAG laser (532 nm second harmonic output) pumping a tunable dye laser generating output near 615 nm, which is then frequency doubled using a BBO crystal. The laser beam is softly