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The effect of fuel oxidation on plasma decay in combustible mixtures excited by high-voltage nanosecond repetitively pulsed discharge

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ABSTRACT

The results of the experimental study of high-voltage nanosecond repetitively pulsed discharge afterglow in propane:O_2, ethane:O_2 and H_2:O_2 mixtures were presented for room temperature and pressures from 2 to 6 Torr. Time-resolved electron density during the plasma decay was measured using a microwave interferometer for initial electron densities in the range between 3×10^{11} and 3×10^{12} cm^{-3} and the effective recombination coefficients were obtained. In hydrocarbon-containing mixtures, the rate of plasma decay varied nonmonotonously with the oxidation degree increase. The effective recombination coefficient (i) peaked when the amount of intermediate species was expected to be high and (ii) was independent of the number of voltage pulses (the effect of saturation) when oxidation was complete. In H_2:O_2 mixtures, the rate of plasma decay and the effective recombination coefficient increased monotonously with increasing oxidation degree. It was shown that plasma decay in completely oxidized fuel:O_2 mixtures is close to that in water vapor. It was suggested that the nonmonotonous behavior of the effective recombination coefficient during plasma decay in hydrocarbon:O_2 mixtures is explained by the production of some hydrocarbon intermediates in the oxidation processes that can favor plasma decay. The possible mechanisms of plasma decay acceleration in the oxidized mixtures were discussed.
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1. Introduction

The improvement of ignition and combustion by non-equilibrium discharge plasmas has received increased attention in recent years due to promising applications of plasma-assisted ignition and plasma-assisted combustion [1–7]. It was demonstrated experimentally and numerically that the use of non-equilibrium discharges can lead to ignition delay reduction, ignition temperature reduction, flame stabilization and increase of flammability limits in various combustible mixtures. Therefore, nowadays plasma-assisted ignition and combustion are considered a promising application of low-temperature non-equilibrium plasmas under various conditions including conditions of high-speed flows and conditions similar to automotive engines [4,6].

To understand the mechanism of plasma-assisted combustion and to numerically simulate the effect of non-equilibrium discharge plasmas on ignition and combustion the chemical kinetic pathways excited by the plasmas and corresponding reaction rates must be studied. Kinetic mechanisms required for plasma combustion modeling are easier to investigate for high gas temperatures when combustion kinetics is well understood and validated. It is much more difficult to study the effect of non-equilibrium discharge plasmas on oxidation and ignition processes at temperatures below the self-ignition threshold when ignition does not occur in the absence of discharge plasma. At low gas temperatures, oxidation processes have been studied in nanosecond repetitively pulsed discharges [8–14]. Plasma activated oxidation was considered for H_2 [8], alkanes from methane to decane [9,10,13,14] and ethylene [11,12]. In these studies, species and temperature measurements were performed using emission and absorption spectroscopy and gas chromatography. Obtained data allowed validation of the kinetic processes that play an important role in plasma-assisted oxidation and plasma-assisted ignition at low gas temperatures.

Previous studies of plasma activated oxidation considered the effect of discharge plasma on oxidation processes. There is also the effect of oxidation on the properties of discharge plasmas. Fuel oxidation leads to a change in the composition of neutral species in

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combustible mixtures. This can influence discharge development, ion composition, excitation degree and other important properties of plasma. The purpose of this work was to study the effect of oxidation on plasma decay after a nanosecond repetitively pulsed discharge. Time-resolved electron density measurements were made in the discharge afterglow to obtain information about the variation in plasma lifetime and the mechanisms of plasma decay for various numbers of discharge pulses leading to partial or complete fuel oxidation.

2. Experiment

The experimental setup and the method used to measure electron density in the afterglow of a high-voltage nanosecond repetitively pulsed discharge have been described in detail elsewhere [9,15]. The discharge was initiated and developed in a quartz tube with an inner diameter of 47 mm. The high-voltage electrode was cone-shaped (angle 60°) and the low-voltage electrode was a grounded ring. The distance between the electrodes was 20 cm. We studied propane:O_2, ethane:O_2 and H_2:O_2 plasmas at gas room temperature and pressures from 2 to 6 Torr. Some experiments were also conducted in pure gases. The discharge was ignited by voltage pulses with an amplitude of 12 kV in the cable. The amplitude of the pulses applied to the high-voltage electrode was 24 kV; twice the voltage in the cable. The wave impedance of the co-axial cable was 50 Ω. The pulse duration at half-height and pulse rise time were 25 ns and 5 ns, respectively. The pulse frequency varied between 10 and 20 Hz. This variation did not affect obtained results. The voltage amplitude, the shape of the pulses and the deposited energy were measured using a back-current shunt.

Electron density in the decaying discharge plasma was measured by a microwave interferometer with a reference wave frequency of 94 GHz (a wavelength of 3 mm). The critical electron density for this frequency is $n_{e\ cr} = 1.2 \times 10^{14}\ \mathrm{cm}^{-3}$, much higher than the electron density for the conditions studied. The power of the diagnostic microwave beam was in the range 1–3 mW. In this case, the increase in the electron temperature due to microwave heating was negligible (less than 1 K).

To measure electron density an incident microwave signal crossed the plasma volume in the middle of the discharge gap perpendicularly, reflected from the angle reflector, crossed the plasma volume again and returned to the waveguide. The incident and reflected waves formed a standing wave in the waveguide. The phase shift of the reflected wave was a function of the electron density in the plasma. In the simplest case of a plane uniform plasma layer, the relation between the phase shift, $\Delta\varphi(t)$, and the electron density, $n_e(t)$, is expressed as [16]

$$n_e(t) = \frac{2\varepsilon_0 mc}{e^2}\frac{\omega\Delta\varphi(t)}{l}, \qquad (1)$$

where m is the electron mass, ε_0 is the permittivity of vacuum, c is the speed of light in vacuum, e is the elementary charge, ω is the angle frequency and l is the path length of the probing wave in the plasma. Eq. (1) is fulfilled for $n_e << n_{e\ cr}$ and $\nu_m << \omega$, where ν_m is the frequency of the electron momentum transfer in collisions with neutral particles. These inequalities were valid under the conditions considered. The absolute uncertainty of the electron density measurements was around 30%. The obtained time-resolved electron density was averaged over the path length of the probing wave in the plasma and over the incident beam diameter (equal to a horn antenna diameter of 35 mm).

3. Measured results and discussion

Figure 1 shows the measured fraction of the deposited energy (the percentage of incident pulse energy) and the specific (per one neutral particle) deposited energy versus the number of voltage pulses. The specific deposited energy was determined considering the variation in the gas number density due to oxidation processes. This variation was estimated from measured gas pressure while neglecting gas temperature variation. The specific energy input per single voltage pulse was between 0.002 and 0.007 eV per one neutral particle. This quantity depended on gas pressure, gas composition and oxidation degree. In particular, the specific deposited energy decreased with the pressure increase. From fig. 1(b), the deposited energy in the propane:O_2 mixture is twice as large as that in the H_2:O_2 mixture. In addition, in the propane:O_2 mixture, the deposited energy percentage and the specific deposited energy demonstrate a minimum at a pulse number $\sim 10^3$. Specific energy does not vary with increasing oxidation degree in the H_2:O_2 mixture.

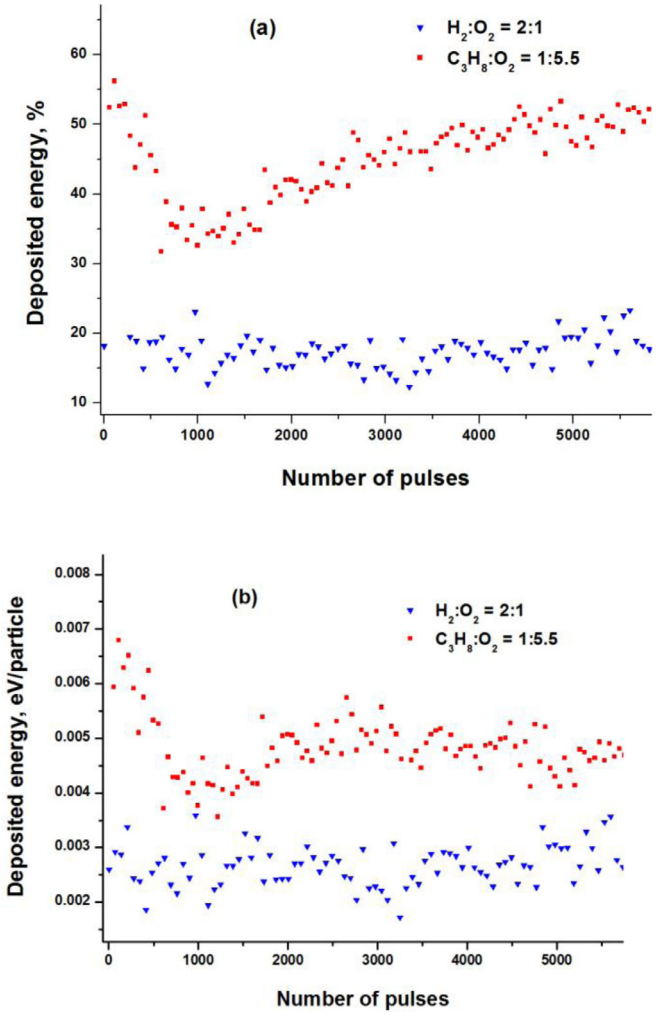

Fig. 1. Deposited energy percentage (a) and specific deposited energy (b) in the nanosecond discharge afterglow in propane:O_2 and H_2:O_2 mixtures for 2 Torr as a function of the number of voltage pulses.

Our estimates show that rapid gas heating during one discharge pulse and its afterglow and gas heating due to energy release in the oxidation processes did not exceed 15 K. In addition, the thermal energy had no time to accumulate in the discharge tube for a large number of voltage pulses because the energy deposited by the discharge pulses was transferred to the discharge tube surface in the time interval between the pulses due to thermal conductivity. Here, the characteristic time of gas cooling in the discharge tube was several orders of magnitude shorter than the time be-

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