Ignition of Ethylene–Air and Methane–Air Flows by Low-Temperature Repetitively Pulsed Nanosecond Discharge Plasma

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Abstract—This paper presents results of low-temperature plasma-assisted combustion experiments in premixed ethylene–air and methane–air flows. The plasma was generated by high-voltage, nanosecond pulse duration, high repetition rate pulses. The high reduced electric field during the pulse allows efficient electronic excitation and molecular dissociation, thereby generating a pool of chemically active radical species. The low duty cycle of the repetitively pulsed discharge improves the discharge stability and helps sustain diffuse, uniform, and volume filling nonequilibrium plasma. Plasma temperature was inferred from nitrogen second positive band system emission spectra and calibrated using thermocouple measurements in preheated flows (without plasma). The experiments showed that adding fuel to the air flow considerably increases the temperature in the plasma, up to $\Delta T = 250$ °C–350 °C. On the other hand, adding fuel to nitrogen flow at the same flow and discharge conditions resulted in a much less pronounced plasma temperature rise, only by about $\Delta T = 50$ °C. This shows that temperature rise in the air–fuel plasma is due to plasma chemical fuel oxidation reactions initiated by the radicals generated in the plasma. In a wide range of conditions, generating the plasma in air–fuel flows resulted in flow ignition, flameholding, and steady combustion downstream of the discharge. Plasma-assisted ignition occurred at low air plasma temperatures, 100 °C–200 °C, and low discharge powers, ~100 W (~1% of heat of reaction). At these conditions, the reacted fuel fraction is up to 85%–95%. The present results suggest that the flow temperature rise caused by plasma chemical fuel oxidation results in flow ignition downstream of the plasma.

Index Terms—Absorption spectroscopy, emission spectroscopy, ignition, low-temperature plasma, nanosecond pulses.

I. INTRODUCTION

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VER THE last few years, considerable progress has been made in studies of ignition and flame stabilization by low-temperature nonequilibrium plasmas (e.g., see recent reviews [1], [2] and references therein). In particular, recent experiments on the ignition of premixed hydrocarbon–air flows using transverse RF discharge plasma and nanosecond duration repetitively pulsed plasma demonstrated that large-volume ignition can be produced at plasma temperatures significantly lower than the autoignition temperature, by at least 300 °C–400 °C

[3]–[6]. These experiments also showed that low-temperature plasma ignition and flameholding can be achieved at significantly lower pressures and higher flow velocities compared to capacitor spark and dc arc ignition [3], in a wide range of equivalence ratios and at a relatively modest plasma power budget (a few percent of the fuel heating value) [4]–[6]. Experiments on ignition of nonflowing preheated hydrogen–air and hydrocarbon–air mixtures by a single-pulse fast ionization wave discharge [7], [8] demonstrated that ignition delay time can be substantially reduced compared to autoignition at the same temperature. Finally, repetitively pulsed nanosecond duration plasma has been shown to stabilize lean premixed atmospheric pressure propane–air flames, increase the flame blow-off velocity, and expand the flammability limits, at a very small plasma power budget (~0.1% of the burner power) [9]–[12]. The results also suggest that chemically active species generated in the plasma, such as oxygen atoms and OH radical, may play a critical role in the kinetics of low-temperature plasma fuel oxidation and ignition [10], [13], [14]. However, detailed kinetic mechanism of nonequilibrium plasma-assisted ignition remains not well understood.

These recent advances suggest the possibility of developing an energy-efficient nonequilibrium plasma ignition and flame stabilization method, which could be used both at low pressures and high flow velocities, as well as in fuel lean mixtures, i.e., when common ignition approaches are ineffective. In particular, our recent work [6] demonstrated the use of high-voltage, short pulse duration, high pulse repetition rate discharge for this purpose. This discharge offers two critical advantages over dc, ac, RF, and microwave discharge plasmas. First, the short pulse duration in this type of discharge greatly improves the plasma stability. The pulse duration, a few tens of nanoseconds, is much shorter than the characteristic time for the ionization instability development and glow-to-arc transition $\sim 10^{-3} - 10^{-4}$ s [15]. This makes it possible to operate the discharge at much higher pressures and power loadings compared to other types of nonequilibrium plasmas. Also, the reduced electric field, $E/N$, during the high voltage pulses results in efficient ionization, electronic excitation, and dissociation of molecular species by electron impact, the rates of which have strong exponential dependence on $E/N$ [15]. This may result in generation of large amounts of active radical species at a relatively low plasma power budget. Previously, this type of discharge has also been successfully used by our group for generating uniform plasmas and sustaining high levels of ionization in high-speed,
low-temperature MHD flow control experiments [16], [17], and for singlet delta oxygen generation in high-pressure discharges [18]–[20].

The objective of this paper are: 1) to expand the scope of our previous work on pulsed plasma-assisted combustion [6] using a higher voltage, shorter pulse duration pulsed plasma generator; 2) to obtain systematic measurements of plasma temperature, burned fuel fraction, and flame temperature in a wide range of parameters; 3) to determine whether steady-state flameholding and combustion are achieved at these conditions; and 4) to provide insight into the mechanism of low-temperature plasma-assisted ignition.

II. EXPERIMENTAL

The experiments have been conducted at the Ohio State University high-speed flow plasma combustion facility [3]–[6]. The schematic of the facility is shown in Fig. 1. A premixed flow of air and gaseous hydrocarbon fuel (methane or ethylene) enters the 5 cm × 1 cm rectangular cross section, 31 cm long test section through a 6 ft long, 1-in diameter gas supply line. The facility can use either room air or cylinder air. Spring-loaded shutoff valve and a flash arrester installed in the 1/4 in. diameter fuel supply line enable quick flow shutoff and prevent the flame propagation into the fuel cylinder. The air and fuel mass flow rates are determined by measuring pressures upstream of the sonic choke plates (inserts with pinholes of known area) placed in the air and fuel delivery lines. Downstream of the test section, the flow is diluted with atmospheric air through a vent valve to prevent further combustion in the vacuum system and in the dump tank (see Fig. 1). The 1200 ft³ dump tank is pumped out using an Allis-Chalmers 1900 ft³/min rotary vane vacuum pump. The test section static pressure ranges from 40 torr to about 0.5 atm. The mass flow rate through the test section can be varied from below 1 to 12 g/s. The test section pressure and the mass flow rate can be varied independently. This makes the experimental facility suitable for combustion studies both in high-speed, low-pressure and in low-speed, intermediate pressure flows.

The test section, made of steel, is shown in greater detail in Fig. 2. The flow enters the rectangular cross section test section through a 1/2-in long ceramic honeycomb flow straightener (300 holes per square inch), which also serves as an additional flashback arrester, and passes between two electrode blocks, as shown in Fig. 2. Each of the two 5 cm × 4 cm rectangular electrode blocks are manufactured of macor ceramic and is flush mounted in the top and bottom test section walls, as shown in Fig. 2. The copper electrode plates are placed into recesses machined in the electrode blocks. The electrodes are rounded at the edges to prevent high electric field concentration and “hot spot” formation in the plasma near the edges. The electrodes are separated from the flow by 1/16 in. thick macor ceramic plates, as shown in Fig. 2. To prevent corona discharge formation in the air pockets between the macor block, the copper electrodes, and the ceramic plates, this space is filled by a self-hardening dielectric compound (silicon rubber). The 2-mm diameter copper electrode leads are soldered to the electrode plates and insulated from the grounded test section by 10 mm outer diameter cylindrical sleeves made of macor ceramic, as shown in Fig. 2. The space between the leads and the sleeves is also filled by the silicon rubber. Two macor ceramic inserts are also flush mounted in the side walls of the test section, as shown in Fig. 2, to prevent the discharge between the high voltage electrode and the grounded test section. The main objective of this design was to confine the discharge plasma to the area between the ceramic plates on top and bottom and between the ceramic side wall inserts, without extending to the steel walls of the test section. Striking a discharge between the high voltage electrode and a test section wall would result in arc filament and hot spot formation in the plasma. In the absence of the plasma, the flow temperature in the test section is monitored by a thermocouple inserted through a port in the top of the test section, as shown in Fig. 2.

Four stepped cylinder BK-7 glass windows are used to provide optical access to the discharge region through 10 mm diameter circular holes machined in the test section and in the side wall inserts, as shown in Fig. 2. During the operation, the windows are held in place by atmospheric pressure. If
the test section pressure exceeds atmospheric pressure during ignition, the windows would be pushed out, thereby reducing the pressure overshoot. Two additional rectangular windows, also made of BK-7 glass, are located approximately 1 cm downstream of the discharge region (see Fig. 2). Finally, static pressure/flow sampling port is located at the end of the test section, as shown in Fig. 2.

The electrode blocks shown in Fig. 2 are connected to a Chemical Physics Technologies custom-designed high-voltage (up to 20–25 kV peak), short pulse duration (~20–30 ns), high repetition rate (up to 50 kHz) pulsed plasma generator. The use of this type of pulsed power supplies to generate repetitively pulsed, low-temperature, uniform plasmas for MHD flow control, plasma-assisted combustion and singlet delta oxygen generation is discussed in our recent publications [16]–[20]. The pulses used in the present experiments has active cooling, which enables its steady-state operation at the pulse repetition rate of 50 kHz. The top electrode is connected to the high voltage output of the pulser, and the bottom electrode is grounded. During the pulser operation, current and voltage in the pulsed discharge are measured using a Tektronix P6015 A high voltage probe and a low-capacitance resistive current probe. The current and voltage waveforms are analyzed by a 1 GHz LeCroy WavePro 7100 A digital oscilloscope.

Optical diagnostics used in this paper included visible emission spectroscopy and Fourier transform infrared (FTIR) absorption spectroscopy. For the time-averaging visible emission spectroscopy measurements, we used an Optical Multichannel Analyzer with a Princeton Instruments intensified charge-coupled device (CCD) array camera and a Spectra Physics 0.5 m monochromator with a 1200 g/mm grating. Time-resolved visible emission was measured using an Acton Research UV spectrometer equipped with a photomultiplier (spectral resolution of up to 0.1 nm). The emission spectra of the plasma were taken through the downstream circular window, while emission spectra of the flame were taken through the rectangular window downstream of the discharge region (see Fig. 2). The FTIR absorption spectra of the combustion products were taken with a Biorad 175 C dynamic alignment FTIR spectrometer with liquid nitrogen cooled InSb detector spectrometer. For this, the flow was sampled through the static pressure/flow sampling port at the downstream end of the test section (see Fig. 2) into a 17.5-cm long cylindrical glass absorption cell with two CaF₂ windows placed into an absorption compartment of the Fourier transform (FT) spectrometer (see Fig. 1). Before sampling the flow, the absorption cell and the supply lines are evacuated using a small vacuum pump. Then the sample is drawn off until the absorption cell and the test section pressures equilibrate, after which the absorption cell shut-off valve is closed (see Fig. 1). In the present experiments, absorption spectra are recorded after the sample temperature reached room temperature. That is, a small flow sample quickly cools off onto its way to the absorption cell. The absorption spectra are measured at a resolution of 0.5 cm⁻¹ using an internal source (globar) of the FT spectrometer. All measurements discussed in this paper have been done using dry air–fuel mixtures.

III. RESULTS AND DISCUSSION

Fig. 3 shows typical voltage and current waveforms produced by the pulser connected to the two electrodes shown in Fig. 2, in air flow at $P = 70$ torr and mass flow rate of $\dot{m} = 0.8$ g/s (flow velocity of $u = 14.7$ m/s). The pulse peak voltage and current are approximately 16 kV and 60 A, respectively, with the voltage pulsewidth at half maximum of about 30 ns. Note that in the present experiments the peak pulse voltage is nearly double the voltage achieved in our previous work using a passively cooled pulsed plasma generator, 9 kV [6], while the pulse rise time is significantly shorter, approximately 30 ns versus 50 ns in [6]. At the baseline conditions, i.e., at test section pressure and temperature of $P = 0.1$ atm, $T = 300$ K, the estimated peak reduced electric field in the plasma generated by the pulse is $E/N \sim 70 \cdot 10^{-16}$ V · cm². This upper bound estimate
Fig. 3. (Left) Typical single pulse voltage and current waveforms and (right) a high voltage pulse sequence at \( \nu = 50 \text{ kHz} \). Air, \( P = 70 \text{ torr} \), \( \bar{m} = 0.8 \text{ g/s} \) \((u = 14.7 \text{ m/s})\).

![Voltage, kV](#)

![Current, 10 A](#)

![Time, nsec](#)

![Voltage, kV](#)

![Time, \mu sec](#)

![20 \mu sec](#)

\[ N_2 \text{ emission:} \]

\[ \text{plasma temperature} \]

\[ \text{CH emission:} \]

\[ \text{flame temperature} \]

Fig. 4. (Left) Photographs of the repetitively pulsed discharge plasma (visible through the circular windows in the test section) and (right) of the flame extending downstream of the discharge section. Ethylene-air flow, \( \Phi = 1 \), \( P = 70 \text{ torr} \), \( \bar{m} = 0.8 \text{ g/s} \) \((u = 14.7 \text{ m/s})\), \( \nu = 40 \text{ kHz} \). The flow is left to right. The arrow shows the windows used for the plasma and the flame temperature measurements.

does not take into account the voltage fall across the plasma sheaths and the ceramic plates covering the electrodes. The pulse energy coupled to the flow, calculated from the voltage and current waveforms at these conditions (see Fig. 3), was 2.2 mJ, compared to about 6 mJ in our previous work [6]. The uncertainty of the pulse energy measurements and the effect of the parasitic phase shift between the measured voltage and the current signals were determined by measuring the voltage and current pulses at \( P = 1 \text{ atm} \), when no breakdown was produced between the electrodes. In this case, the pulse energy calculated from the voltage and current waveforms (i.e., the uncertainty in the measured pulse energy) was approximately 0.2 mJ.

Pulse energies measured in air as well as in stoichiometric ethylene-air and methane-air mixtures, at flow rates of \( \bar{m} = 0.8-1.8 \text{ g/s} \) and pressures of \( P = 70-100 \text{ torr} \) were similar, in the range between 1.7 mJ and 2.3 mJ. At these conditions, the average load matching coefficient, defined as the difference between the forward and the reflected pulse energies divided by the forward pulse energy was approximately 25\%, at the average forward pulse energy of about 8 mJ. The forward and the reflected energies were estimated as the positive and the negative contributions to the integrand in the energy integral over the pulse, \( \int U(t)I(t)dt \), respectively, where \( U(t) \) and \( I(t) \) are the voltage and current waveforms shown in Fig. 3.

Fig. 3 also shows several voltage pulses generated at the pulse repetition rate of \( \nu = 50 \text{ kHz} \), at the same flow conditions. From Fig. 3, it can be seen that at this pulse repetition rate the voltage duty cycle is extremely low, \( \approx 30 \text{ ns}/20 \mu s \approx 1/1000 \). The high reduced electric field during the pulses makes possible efficient ionization and dissociation of molecular species by electron impact, the rates of which have strong exponential dependence on \( E/N \) [15]. On the other hand, the short pulse duration and the low duty cycle greatly improve the plasma stability. That is, the pulse duration, \( \approx 30 \text{ ns} \), is much shorter than the characteristic time for the ionization instability development, \( \approx 10^{-3} - 10^{-4} \text{ s} \) [15].

To prevent overheating of the pulsed electrode blocks, the pulser run time was limited to 1–2 s in the present experiment. This time was sufficient to take visible emission spectra of the plasma and to draw a sample of the flow into the absorption cell of the FTIR. The experiments have been conducted using methane and ethylene fuels, at test section pressures of \( P = 70-100 \text{ torr} \), mass flow rates of \( \bar{m} = 0.8-2.4 \text{ g/s} \), and pulse repetition rates of \( \nu = 50 \text{ kHz} \). In the entire range of
experimental conditions, the repetitively pulsed plasma appeared diffuse and stable, occupying a volume of approximately 20 cm$^3$. These visual observations are consistent with plasma images taken by an intensified CCD camera with the gate of 300 ns, which show the uniform distribution of the plasma emission [13]. At the conditions when ignition was achieved in the test section, a flame originated in the plasma and extended downstream through the test section, visible through the rectangular optical access window (see Fig. 4). After ignition, the flame in the test section remains steady as long as the discharge is on. Turning the discharge off results in flame extinguishment, since the test section does not have a flameholder.

Visible emission spectra of the plasma (partially rotationally resolved $0 \rightarrow 2$ band of the $\text{N}_2(\text{C}_3^v \quad \Pi_u \rightarrow \text{B}_3^2 \quad \Pi_g)$ band system) have been used to infer the rotational temperature in the plasma. For this, synthetic spectrum has been used, using nitrogen molecular constants [21], rotational like intensities [22], and the experimentally measured slit function of the spectrometer. This method of temperature inference has to be used with caution, since the rotational relaxation time at the baseline conditions, test section pressure and temperature of $P = 0.1$ atm, $T = 300 \text{ K}$ is $\tau_{\text{rot}} = 12 \text{ ns}$ [23], which is comparable with the radiative lifetime of the $\text{N}_2(\text{C}_3^v \quad \Pi_u \nu = 0)$ state, 38 ns [24]. For this reason, this temperature inference needs to be calibrated. In the present experiment, calibration has been done by comparing the synthetic and the experimental $\text{N}_2(\text{C}_3^v \quad \Pi_u \rightarrow \text{B}_3^2 \quad \Pi_g)$ emission spectra measured in air preheated by an in-line flow heater up to $T = 20-180 \text{ °C}$, at $P = 70 \text{ torr}$ and $\dot{m} = 1.0 \text{ g/s}$. In these measurements, both pulse repetition rate and time-averaged discharge power were low, $\nu = 5 \text{ kHz}$ and $2.3 \text{ mJ} \cdot 5 \text{ kHz} \approx 12 \text{ W}$, respectively. At these conditions, the estimated flow temperature rise in the plasma was also small, $\approx 25 \text{ °C}$.

Fig. 5 compares plasma temperatures inferred from the nitrogen emission spectra with thermocouple measurements in the absence of the plasma. It can be seen that the agreement between the two temperatures is very good, within the uncertainty of the temperature inference, $\pm 25 \text{ K}$. This demonstrates applicability and accuracy of this temperature measurement method at the present conditions. Note that the plasma temperature has been measured in the discharge region (as shown in Figs. 2 and 4), not in the flame region downstream, since $\text{N}_2(\text{C}_3^v \quad \Pi_u \rightarrow \text{B}_3^2 \quad \Pi_g)$ emission in that region becomes very weak. Fig. 6 shows the experimental and the synthetic $\text{N}_2(\text{C}_3^v \quad \Pi_u \rightarrow \text{B}_3^2 \quad \Pi_g)$ emission spectra in a stoichiometric ethylene–air mixture, at $P = 70 \text{ torr}$, $\dot{m} = 0.8 \text{ g/s} (u = 14.7 \text{ m/s})$, and $\nu = 40 \text{ kHz}$, when ignition was achieved and flame was generated downstream of the plasma, as can be seen in Fig. 4. At these conditions, the inferred rotational temperature in the plasma is $T = 630 \text{ °C} \pm 50 \text{ °C}$. Fig. 7 shows the time-resolved $\text{N}_2$ second positive band emission signal from the air plasma (at 375.5 nm) at $P = 70 \text{ torr}$ and $\dot{m} = 0.8 \text{ g/s} (u = 14.7 \text{ m/s})$, for two different pulse repetition rates, $\nu = 20$ and 50 kHz. It can be seen that nitrogen emission is modulated at the pulse repetition rate, reaching maximum during the high-voltage pulses and partially decaying between the pulses. Note that the decay time of the $\text{N}_2(\text{C}_3^v \quad \Pi_u \rightarrow \text{B}_3^2 \quad \Pi_g)$ emission at

![Fig. 5. Comparison of preheated flow temperatures inferred from $\text{N}_2$ second positive spectra with thermocouple measurements. Air, $P = 70 \text{ torr}$, $\dot{m} = 1.0 \text{ g/s} (u = 18.4 \text{ m/s}), \nu = 5 \text{ kHz}$, discharge power 12 W.](image1)

![Fig. 6. Experimental and synthetic $\text{N}_2$ emission spectra (second positive band system) of the plasma. Ethylene–air mixture, $\Phi = 1$, $P = 70 \text{ torr}$, $\dot{m} = 0.8 \text{ g/s} (u = 14.7 \text{ m/s}), \nu = 40 \text{ kHz}$, with flame observed in the test section. Best fit rotational temperature is $T = 900 \text{ K} \pm 50 \text{ K}$.](image2)

![Fig. 7. Time-dependent $\text{N}_2$ positive system emission signal (375.5 nm) in the pulsed discharge plasma at two different pulse repetition rates, (top) 50 kHz and (bottom) 20 kHz. Air, $P = 70 \text{ torr}$, $\dot{m} = 1.0 \text{ g/s} (u = 14.7 \text{ m/s})$.](image3)
the present conditions is of the order of 100 μs, which is 
approximately three orders of magnitude longer than the 
radiative lifetime of the N2(C3Πu) state. This demonstrates that the 
C3Πu state of nitrogen continues to be populated between the 
high voltage pulses.

For the flame detection, both visual observations and visible 
emission spectra have been used. As in our previous work 
[3]-[6], CH 4300 Å band system emission, CH(A2Σ→X2Π)), has been used as a flame indicator. Fig. 8(a) shows 
time-resolved CH emission at 431.4 nm from a stoichiometric ethylene-air mixture in the flame region downstream of the 
plasma, measured at P = 70 torr, \( \dot{n} = 0.8 \) g/s (u = 14.7 m/s) and \( \nu = 20-40 \) kHz. It can be seen that at \( \nu = 40 \) kHz, 
when the flame is visually detected in the test section, CH 
emission is relatively steady and does not exhibit modulation 
observed in the N2 emission (compared with Fig. 7). This 
suggests that at these conditions combustion downstream of the 
plasma is self-sustained. Decreasing the pulse repetition 
rate to \( \nu = 30 \) kHz reduces CH emission intensity and makes 
it much less steady in time, exhibiting sudden increases and 
suggesting unstable combustion regime triggered by individual 
high-voltage pulses [see Fig. 8(a)]. Finally, at \( \nu = 20 \) kHz, 
CH emission nearly disappears and combustion appears to be 
not sustained. Since the flow residence time in the discharge 
section at \( u = 14.7 \) m/s is approximately 2.7 ms, these results 
suggest that self-sustained combustion downstream of the 
discharge is achieved after the combustible mixture is excited by \( \sim 110 \) high voltage pulses. Time-resolved emission 
measurements at \( \nu = 30 \) kHz and at different flow velocities, 
\( u = 15-44 \) m/s, shown in Fig. 8(b), is consistent with these 
results. Increasing the flow velocity from the baseline value of 
\( u = 14.7 \) m/s to \( u = 44 \) m/s, i.e., reducing the number of pulses 
exciting the combustible mixture from \( \sim 110 \) to \( \sim 35 \) pulses 
considerably reduces CH emission intensity and makes it much 
less steady in time, again suggesting an unstable combustion 
regime.

The R-branch of the partially rotationally resolved 
CH(A2Σ→X2Π) emission spectra, (0,0) and (1,1) 
bands, have been used to infer the flame temperature. This 
method has been previously used to infer hydrocarbon 
flame temperatures [25, 26]. In particular, analysis of high-
resolution CH emission spectra [26] showed no evidence of 
self-absorption, which may considerably affect the temperature 
value inferred from partially rotationally resolved spectra. 
Spectroscopic constants of CH molecule, rotational level 
energies, and rotational line intensities were taken from 
[27], [28]. Fig. 9 compares the experimental CH spectrum 
in a stoichiometric ethylene-air mixture, at \( P = 70 \) torr, 
\( \dot{n} = 0.8 \) g/s (u = 14.7 m/s), and \( \nu = 50 \) kHz, with the 
synthetic spectrum calculated at the rotational temperature of 
\( T = 1730 \) °C. The flame temperature inferred from fitting the 
two spectra is \( T = 1700 \) °C ± 300 °C.

Fig. 10 shows typical FT absorption spectra of the stoichiometric ethylene–air flow sampled downstream of the discharge 
section at \( P = 70 \) torr, \( \dot{n} = 1.0 \) g/s (u = 18.4 m/s), and \( \nu = 50 \) kHz, with and without the plasma. At these conditions, 
turning on the repetitively pulsed discharge resulted in flow 
ignition, with a flame extending through the test section (such
as shown in Fig. 4). When ignition was achieved, CH 4300 Å band system emission was also detected from the flame downstream of the plasma region. From Fig. 10, it can be seen that the use of the repetitively pulsed plasma results in a nearly complete disappearance of ethylene and an increase in concentrations of CO, CO₂, and H₂O in the flow. It can also be seen that oxidation of ethylene does not result in its conversion to acetylene (absorption band between 3230 and 3350 cm⁻¹), which has been observed in fuel-rich mixtures excited by an RF discharge [4].

Fig. 11 shows the effect of the flow velocity in the test section on the temperature in the plasma and on the reacted fuel fraction. These measurements have been done in a stoichiometric ethylene-air flow at the same pressure and pulse repetition rate, P = 70 torr and ν = 50 kHz. Fig. 11 also shows the results of flow temperature measurements in air plasma (without fuel) at the same conditions. Note that the air plasma temperature measured before adding the fuel is quite low, T = 130 °C–230 °C. It can be seen that while both the air flow and the fuel-air mixture temperature decrease with the flow velocity as it increases from u = 14.7 m/s to u = 44 m/s (at \( m = 0.8–2.4 \) g/s), the temperature of the air–fuel mixture is significantly higher, by \( \Delta T = 200 °C–350 °C \). This suggests that the temperature increase may be due to heat generation during plasma chemical fuel oxidation. Note that the measured flow temperature rise of \( \Delta T = 350 °C \) at u = 14.7 m/s and \( \Delta T = 200 °C \) at u = 44 m/s (see Fig. 11) correspond to oxidation of approximately 12% and 7% fraction of ethylene in the plasma, respectively (the heat of combustion reaction for a stoichiometric ethylene–air mixture is 3025 KJ/kg).

The results of Fig. 11 also suggest that when the plasma temperature approaches autoignition temperature (for ethylene–air mixtures, \( T_{\text{auto}} = 570 °C–700 °C \) at P = 70 torr [29]), ignition occurs downstream of the plasma. Note that although complete combustion was achieved only at u = 14.7 m/s (reacted fuel fraction of 95%, see Fig. 11), ignition was produced and flame was detected in the entire range of flow velocities of up to u = 44 m/s, i.e., at the air plasma temperatures as low as T = 120 °C (see Fig. 11). A long and stable flame was detected in the test section even at the conditions when the measured reacted fuel fraction was only a few percent, at u = 44 m/s (see Fig. 11). This suggests that at these conditions most of the fuel
burns downstream of the fuel sampling port (see Fig. 2). At $u = 44$ m/s, the estimated discharge energy loading per unit mass necessary for ignition is $100$ W / $2.4$ g/s $\sim 40$ kJ/kg (or $\sim 0.01$ eV/molecule). This is about $1.3\%$ of the heat of combustion reaction. These results demonstrate that ethylene–air flows can be ignited by the repetitively pulsed plasma at low plasma temperatures, and with little flow preheating by the plasma.

To verify whether the temperature rise in the air–fuel flow, compared to the air flow (see Fig. 11), is indeed due to plasma chemical fuel oxidation rather than relaxation of excited species generated in the air plasma, the entire series of measurements shown in Fig. 11 was repeated after air was replaced with nitrogen. The results are summarized in Fig. 12. It can be seen that in this case the temperature rise in the nitrogen–fuel flow, compared to the nitrogen only flow, is much less significant, only about $\Delta T = 50$ °C. This is most likely due to faster relaxation of energy stored in the electronically and vibrationally excited nitrogen molecules in the presence of ethylene. From Fig. 12, one can also see that at these conditions the reacted fuel fraction does not exceed $5\%$, which is close to the uncertainty of the present FTIR spectroscopy measurements. Summarizing, the results shown in Figs. 11 and 12 demonstrate that: 1) temperature rise in the air–fuel plasma indeed occurs due to energy release during plasma chemical fuel oxidation; and 2) direct fuel dissociation by electron impact is a relatively minor reaction channel.

Fig. 13 summarizes the results of plasma-assisted combustion measurements in ethylene–air mixtures at $P = 70$ torr, $\dot{\bar{v}} = 1.0$ g/s ($u = 18.4$ m/s), and $\nu = 50$ kHz, for different equivalence ratios. From Fig. 13, it can be seen that both the flow temperature and the reacted fuel fraction are highest at near stoichiometric conditions, $T = 490$ °C and $72\%$–$87\%$, respectively.

Fig. 14 shows typical FT absorption spectra of the stoichiometric methane–air flow sampled downstream of the discharge section, at $P = 100$ torr, $\dot{\bar{v}} = 1.0$ g/s ($u = 12.9$ m/s), $\nu = 50$ kHz, with and without the plasma. As in ethylene–air, it can be seen that the use of the repetitively pulsed plasma results in a significant reduction of the methane concentration and an increase in concentrations of CO, CO$_2$, and H$_2$O in the flow (compare Figs. 10 and 14). Again, no acetylene absorption between 3230 and 3350 cm$^{-1}$ has been detected. The dependence of the plasma temperature and of the reacted methane fraction on flow velocity and equivalence ratio, measured at $P = 100$ torr and $\nu = 50$ kHz (see Figs. 15 and 16) is similar to the results for ethylene–air mixtures, shown in Figs. 11 and 13. Ignition was achieved and flame was detected in the test section, while significant amount of fuel reacted at flow velocities of $u = 10$–$23$ m/s ($\dot{\bar{v}} = 0.8$–$1.8$ g/s) and air flow temperatures as low as $T = 100$ °C–$200$ °C. Similar to ethylene–air results, adding methane to the air flow results in the temperature increase in the plasma by up to $\Delta T = 250$ °C, which corresponds to oxidation of approximately $8\%$ fraction of methane in the plasma (see Fig. 15). This results in ignition and burning of nearly $90\%$ of the fuel downstream of the plasma, as shown in Fig. 15. At $P = 100$ torr, $\dot{\bar{v}} = 0.8$ g/s ($u = 12.9$ m/s), and $\nu = 50$ kHz, the flame temperature in a stoichiometric methane–air mixture, inferred from the CH emission spectrum, is $T = 1600$ °C ± $300$ °C. The highest flow temperature in the plasma, $T = 430$ °C, and reacted fuel fraction, $75\%$–$88\%$, are also achieved at near stoichiometric conditions (see Fig. 16). Note that the present results are quite different from our previous paper [6], where ignition in methane–air mixtures was not achieved and flame was not detected in the entire range of flow velocities and equivalence ratios, in spite of significant fractions of fuel oxidized.

Comparing the results of the present experiments with our previous measurements [6], in this paper the air plasma temperature (before the fuel is added) is significantly lower than in [6] at the same flow conditions. For example, at $P = 70$ torr, $u = 14.7$ ms, and $\nu = 50$ kHz, the temperature is $T = 230$ °C (see Fig. 11), compared to $T = 370$ °C in [6]. At $P = 100$ torr, $u = 12.9$ m/s, and $\nu = 50$ kHz, the temperature is $T = 200$ °C (see Fig. 15), compared to $T = 390$ °C at a lower pulse frequency of $\nu = 40$ kHz in [6]. Lower plasma temperatures measured in this paper are due to significantly lower pulse energy produced by the new pulser, approximately $2$ mJ versus about $6$ mJ in [6]. However, the range of flow velocities at which ignition is achieved in this paper is considerably wider than in [6], $u = 15$–$45$ m/s versus $u = 15$–$25$ m/s in ethylene–air and $u = 10$–$20$ m/s versus no ignition in methane–air. We believe that the difference is mainly due to a significantly higher pulse voltage (i.e., higher $E/N$) and shorter pulse rise time generated by the new pulsed power supply. This suggests that the use of higher pulse voltage (up to 30–40 kV), higher pulse repetition rate (up to $\nu = 100$ kHz), and shorter pulse duration (a few nanoseconds), such as that generated by a Fast Ionization Dynistor Technology pulse generator [19] would produce low-temperature plasma ignition and flameholding at significantly higher flow velocities, up to $\sim 100$ m/s.

IV. Summary

This paper presents results of plasma-assisted combustion experiments in premixed hydrocarbon–air flows excited by
Fig. 13. Flow temperature in the discharge section and unreacted fuel fraction downstream of the plasma as functions of the equivalence ratio. Ethylene-air, $P = 70$ torr, $m = 1.0$ g/s ($u = 18.4$ m/s), $\nu = 50$ kHz.

Absorption

Air-methane
$P = 50$ torr, $\Phi = 1$, $u=13$ m/s
plasma off
plasma on

CO$_2$
H$_2$O
CH$_4$
CO

Wavenumbers (cm$^{-1}$)

Absorption

photon off
photon on

CH$_4$

Wavenumber (cm$^{-1}$)

Fig. 14. Typical FT absorption spectra of the flow sampled downstream of the discharge section, with and without repetitively pulsed plasma. Methane-air, $\Phi = 1$, $P = 90$ torr, $m = 1.0$ g/s ($u = 13$ m/s), $\nu = 50$ kHz.

Temperature [°C]

P=100 torr, $\Phi=1$

Air
Air-methane

Flow Velocity [m/s]

Unreacted Fuel [%]

Air-methane
$P=100$ torr, $\Phi=1$

Flow Velocity [m/s]

Fig. 15. (Left) Flow temperature in the discharge section and (right) unreacted fuel fraction downstream of the plasma as functions of the flow velocity. Methane-air, $\Phi = 1$, $P = 100$ torr, $\nu = 50$ kHz.

a low-temperature, transverse, repetitively pulsed discharge plasma. The experiments have been conducted with methane and ethylene fuels in a wide range of equivalence ratios and flow velocities. The plasma was generated by high-voltage (16–18 kV), short pulse duration (20–30 ns), high repetition rate (up to 50 kHz) pulses. The high reduced electric field
during the pulse allows efficient electronic excitation and molecular dissociation, thereby generating a pool of chemically active radical species. On the other hand, the low duty cycle of the repetitively pulsed discharge, ~1/1000, greatly improves the discharge stability and helps sustain diffuse, uniform, and volume filling nonequilibrium plasma. Plasma temperature was inferred from nitrogen second positive band system emission spectra and calibrated using thermocouple measurements in flows preheated by an in-line flow heater (without plasma). The experiments showed that adding fuel to the air flow considerably increases the flow temperature in the plasma, up to $\Delta T = 250^\circ$C$-$350$^\circ$C. On the other hand, adding fuel to nitrogen flow at the same flow and discharge conditions resulted in a less pronounced plasma temperature rise of only about $\Delta T = 50^\circ$C. This shows that temperature rise in the air–fuel plasma is due to plasma chemical fuel oxidation reactions initiated by the radicals generated in the plasma.

In a wide range of operating conditions, generating the repetitively pulsed plasma in premixed air–fuel flows resulted in flow ignition and flameholding downstream of the discharge. The flame originated in the plasma, extended downstream through the test section, and remained stable as long as the plasma was on. Flame temperature, up to $T = 1700^\circ$C$\pm 300^\circ$C, was inferred from CH 4300 Å band system emission spectra. Time-resolved visible emission spectroscopy demonstrated that repetitively pulsed nanosecond discharge produces self-sustained combustion downstream of the plasma region. The results show that plasma-assisted ignition occurred at low discharge powers, ~100 W (~1% of heat of reaction), and low air flow temperatures, 100$^\circ$C$-$200$^\circ$C. At these conditions, the reacted fuel fraction, measured by the FTIR absorption spectroscopy, is up to 85%–95%. The present results suggest that temperature rise caused by exothermic plasma chemical fuel oxidation results in flow ignition downstream of the plasma. Both the plasma temperature and the reacted fuel fraction peak at near stoichiometric conditions. Further experiments to provide insight into kinetic mechanism of the low-temperature plasma ignition are currently underway.

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