Plasma-Discharge Stabilization of Jet Diffusion Flames

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***Abstract*—The authors examine three different types of plasma discharges in their ability to stabilize a lifted jet diffusion flame in coflow. The three discharges include a single-electrode corona discharge, an asymmetric dielectric-barrier discharge (DBD), and a repetitive ultrashort-pulsed discharge. The degree of nonequilibrium of this pulsed discharge is found to be higher than that for the DBD. Furthermore, this pulsed discharge causes the most significant improvement in the flame stability. The optimal placement of the discharge electrodes is investigated, and it is found that there is a close relation between this placement and the emission spectra, suggesting use of the emission spectra as a possible indicator of fuel/air mixture fraction. The optimal placement is mapped into mixture-fraction space by use of a fully premixed flame experiment of known mixture fraction. The result shows that the mixture fraction, which corresponds to the optimal placement, is much leaner than that of a conventional lifted jet flame.**

***Index Terms*—Diffusion, discharge, flame stabilization, lifted, nonequilibrium.**

I. INTRODUCTION

THE ISSUE of flame stability is receiving renewed attention in the burning of gaseous hydrocarbons because of the increased demand for high-power/low-emission combustion and the trend toward the utilization of low-grade fuels. Several methods have been used to achieve stabilization in combustion systems, such as pilot flames, oxygen-rich coflows, bluff bodies, and swirling flows. Pilot flames are routinely used to stabilize laboratory scale, nonpremixed [1], [2], and premixed [3]–[5] flames. A pure oxygen coflow surrounding a jet flame was demonstrated to be successful in stabilizing laboratory-scale diffusion flames [6]. Both of these methods add considerable complexity to an overall industrial-scale system. Bluff body and/or swirl mechanisms have been successfully employed in both premixed and partially premixed configurations. These approaches generate a recirculation zone, which preheats the reactants, resulting in increased flame stability [7]–[10]. However, the resulting increased entrainment of high-temperature burned gas into the fresh reacting jet by recirculation and the longer residence time of flow in the reaction region by such swirling motion can also lead to a significant increase in the formation of $\mathrm{NO_X}$.

In this paper, we discuss recent studies on the use of nonequilibrium plasma discharges to enhance the stability of a lifted methane-jet flame. The use of nonequilibrium discharges is unlike those mentioned above in that these discharges create cold radicals in a combustible environment. This is contrasted to the use of pilot flames where the main energy transfer occurs predominantly in the form of thermal energy, a portion of which is lost while local thermal equilibrium is established [11]. Although the translational temperature of the formed radicals is close to room temperature, the electronic and vibrational temperatures can be much higher due in part to higher electron temperatures, potentially increasing the rates of branching reactions. This nonequilibrium plasma-assisted energy mode targeting can be achieved by decreasing the discharge time scale (e.g., short-pulse discharges). When the discharge time is less than the energy-transfer time between electronic and translational modes, a larger fraction of the added energy results in bond breaking, rather than increasing the local gas temperature.

Various methods of achieving nonequilibrium pulsed discharges have been reported in literature for applications related to combustion and/or combustion stabilization. Among them are the dielectric-barrier discharges (DBD) and alternating-current corona discharges, which are known to achieve highly nonequilibrium conditions [12]–[19]. A DBD is an ac-driven discharge that, in most conditions, produces pulsed microstreamers. Okazaki and Nozaki [12] and Cha *et al.* [16] used a DBD to reduce the concentration of $\mathrm{NO_X}$, $\mathrm{SO_X}$, and soot produced by combustion. Starikovskii [15], Mintoussov *et al.* [17], and Starikovskaia *et al.* [18] implemented the nanosecond pulsed streamer discharge characteristic of the DBD to increase flame stability. They observed a reduction in ignition delay time and an increase in the stability limit for a premixed flame. We have reported favorable results of a study of the use of a nonequilibrium ac corona discharge applied to the flame stabilization of a methane jet in an air coflow [19]. Both DBD and corona discharges are relatively simple to integrate into a combustion system and are of relatively low cost. In practice, DBD and corona discharges in air or air–fuel mixtures at high pressure tend to be filamentary, giving rise to bursts of microdischarges, of peak current, and frequency that are not directly controllable. A less studied nonequilibrium discharge is that studied here—an ultrashort-pulse repetitive discharge (USRD). Single repetitive pulses of controlled frequency as high as 100 kHz is found to lead to a peak discharge current that is much higher than those generated by the filamentary bursts

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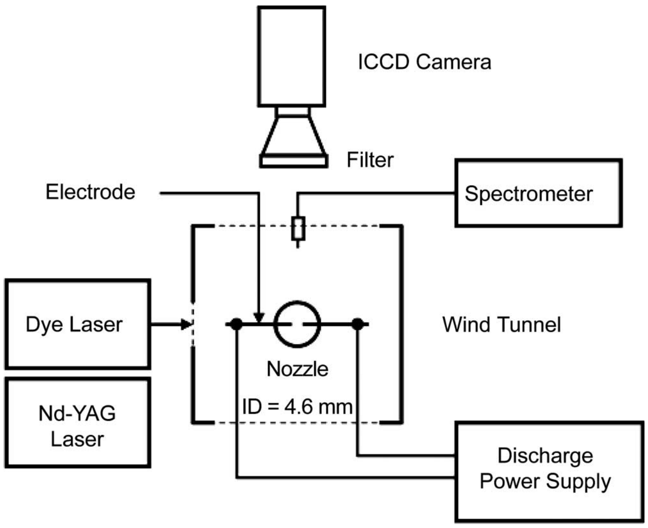


Fig. 1. Schematic of the experimental setup.

in these other discharges. Kim *et al.* showed that the stability limit of the swirl stabilized methane/air-premixed flame could be extended with a USRD beyond the thermochemical lean flammability limit of the methane [20]. Galley *et al.* also observed a stability-limit extension using a USRD for a bluff-body stabilized premixed V-shaped flame [21]. As described by Pancheshnyi *et al.*, the USRD was applied to improve the ignition of a propane/air mixture at high pressure [22]. Lou *et al.* showed evidence for a significant preflame oxidization of methane/air and ethylene/air mixtures when ignited with a USRD [23].

A comparison of the ability of these discharges, i.e., the single electrode ac corona discharge (SECD), DBD, and USRD, to extend flame stability is the main focus of this paper.

The scope of this paper is focused on the stabilization of lifted methane-jet flames. The current understanding of the stabilization mechanism of a natural lifted jet flame is that of a leading edge flame of triplet character for both laminar and turbulent jets, which implies that the flame base is anchored instantaneously on a triple point of three branches where a competition occurs between the flame propagation speed and the local flow velocity [24], [25]. This perspective matches well with our previous observations in which the lifted flame base is located in a flow field where the local flow velocity is two to three times that of the laminar flame speed (S_{L}) [26]. The natural lifted jet flame is considerably more difficult to stabilize at velocities exceeding 2–$3S_{\mathrm{L}}$ of coflow velocity regardless of velocity of the jet [27]. Thus, in this paper, we use coflow velocity and jet velocity as the main metrics against which the degree of improvement of discharge-assisted flame stabilization is evaluated. In addition, we survey the electrode placement, which provides maximum stability to the lifted jet flame.

II. EXPERIMENTAL SETUP

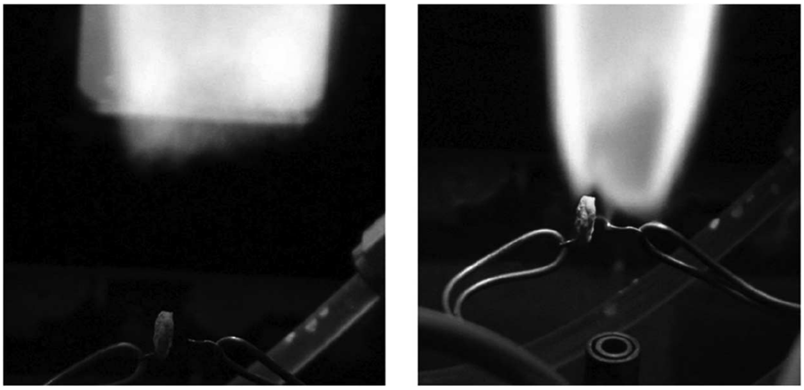
A schematic diagram of the experimental setup is provided in Fig. 1. A lifted methane-jet flame is formed in a vertical wind tunnel that is 30 × 30 cm in its cross section. A nozzle with inner diameter of 4.6 mm is oriented parallel to the flow direction to produce a jet in coflow. Coflow velocity is measured by particle image velocimetry (PIV) while the jet velocity is determined by measuring upstream pressure and volume flow rate using flowmeters. The PIV system consists of a 15-Hz double-exposure interlaced charge-coupled device (CCD) camera (Kodak ES 1.0), a 15-Hz double-pulse second harmonic Nd:YAG laser (Spectra Physics PIV-400), and an alumina-particle seeding system. In addition, an intensified-CCD (ICCD) camera (Princeton instrument PI-MAX) with a 50-kHz maximum gating frequency and 1.5-nm minimum gate width is used to qualitatively visualize the plasma discharge in a time resolved manner. The voltage and current at the electrode are recorded with a 1000 : 1 high-voltage probe (Tektronics P6015A) and Rogowski coil (Pearson Electronics, model 2877), respectively. In addition, we also record spectrally resolved plasma optical emission as a broad survey over a wide spectral window with a relatively course spectral resolution of 0.17 nm using an Ocean Optics (S2000) spectrometer and also at a higher resolution of < 0.1 nm with a SPEX750M spectrometer equipped with a 2000 × 800 pixel CCD camera.

For the DBD and SECD studies, the discharge is powered by an ac power supply with a typical peak-to-peak voltage of $\sim$10 kV and typical frequency range of 25–35 kHz. For the USRD studies, we use a pulsed power supply (FID Technology SU-12) that provides pulses of 6-kV peak voltage (typical), 10-ns pulsewidth, and 15-kHz pulse repetition rate. Opposed, pointed electrode pairs made of platinum (Pt) and tungsten (W) are used for the USRD. A single (powered) platinum electrode (the distant flame serves as a virtual ground) and a pair of platinum (powered)/quartz coated platinum (grounded) electrodes are used for the SECD and DBD studies, respectively. A high sampling frequency photodiode is used to detect the flame's emission and to qualitatively confirm the presence of a flame from which its duty cycle (defined as the fraction of the total time that the flame is ignited) can be estimated.

III. RESULTS AND DISCUSSION

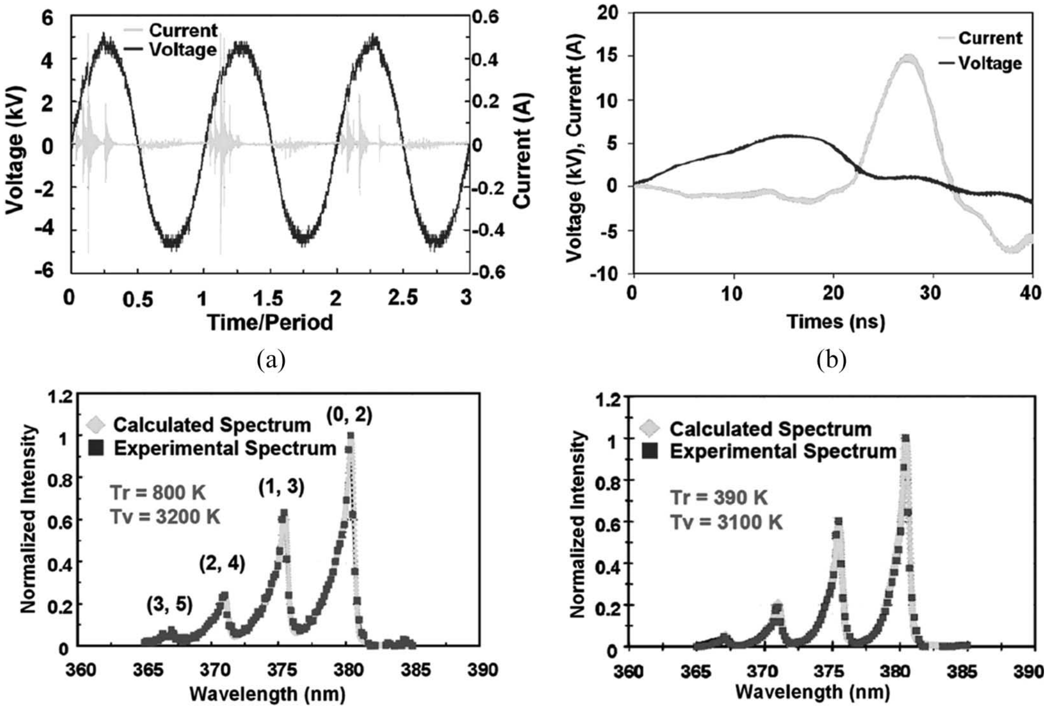
A. Discharge Comparison

In this section, we compare the ability of these three high-pressure nonequilibrium discharges, namely SECD, DBD, and USRD, to improve the flame stability. Fig. 2 shows a representative photograph of a DBD discharge configuration between a bare platinum electrode and a sapphire covered platinum electrode. In Fig. 2(a), in the absence of a discharge, the flame is clearly lifted and is located approximately 50 mm above the electrode pair, which itself is located 46 mm downstream of the jet nozzle. With the discharge initiated, as shown in Fig. 2(b), the flame is pulled down and attached to the plasma in the vicinity of the electrode pair. A close examination of the near-electrode region in Fig. 2(b) shows that there are two distinct discharge kernels: a relatively intense discharge between the electrodes (i.e., the DBD) and also a diffuse discharge from the bare-platinum-electrode tip to the flame base. This secondary discharge is the basis for our single electrode configuration (SECD). We believe that the high-temperature flame itself serves as a virtual electrode as it acts as a large reservoir of charged particles and has a finite bulk capacitance.



(a) (b)

Fig. 2. Observations of discharge-enhanced flame stabilization. (a) Without discharge. (b) With discharge. The flame is lifted OFF in (a) and reattached by discharges in (b). SECD as well as DBD is observed in (b). The electrode position is ten diameters (46 mm) above the nozzle.



(a) (b)

(c) (d)

Fig. 3. (a) Typical voltage and current curve of DBD showing many microdischarges in one cycle. The period is 33 μs. (b) Typical voltage curve of USRD showing only one discharge in one period of a repetition cycle. Second positive vibrational band spectra of nitrogen with $\Delta\nu = 2$ measured in (c) DBD and (d) USRD. Squares represent experimental values. For comparison, diamonds are calculated values at specific rotational and vibrational temperatures.

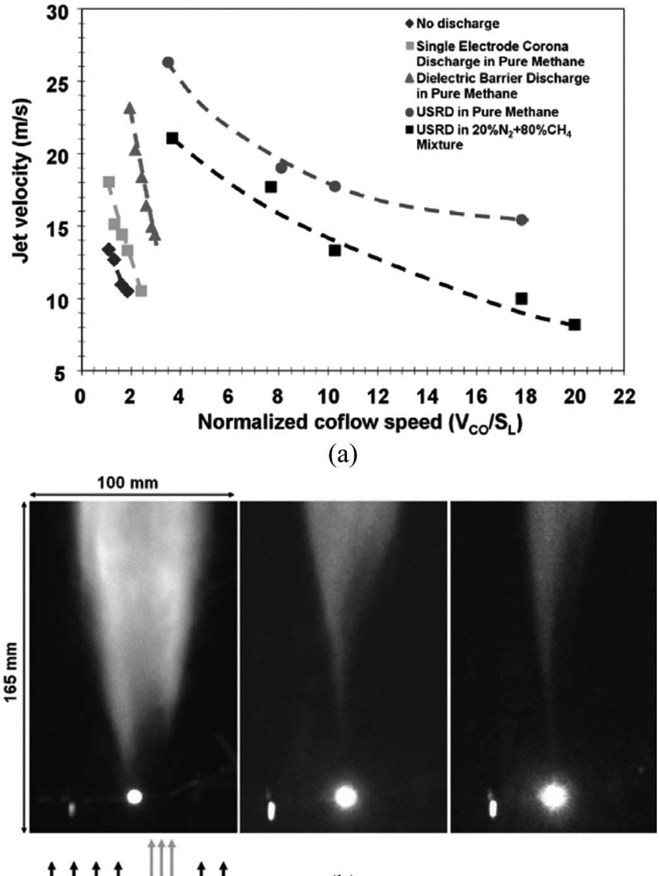
Typical voltage and current traces of the DBD and USRD in quiescent air are illustrated in Fig. 3(a) and (b), respectively. The presence of numerous microdischarges during a single cycle in the DBD case [Fig. 3(a)] is apparent from the current bursts. While each individual discharge has a very short pulsewidth (~10–100 ns), the bursts span an overall timescale ~1–10 s. By comparison, the voltage profile of the USRD is shown in Fig. 3(b). In this case, there is a single ~10-ns pulse at a controllable repetition rate. The peak current of the USRD exceeds 15 A, i.e., approximately 30 times that of any single pulse in the DBD. The higher current in the USRD is in part due to the absence of a dielectric barrier, which terminates the individual discharge current pulses in the DBD due to charge accumulation on the surface of the dielectric. In the absence of a dielectric, the peak discharge current in the DBD configuration is found to increase at least five-fold.

Fig. 3(c) and (d) shows representative discharge emission spectra from the second positive system of molecular nitrogen $(C^3\prod_u \rightarrow B^3\prod_g)$ with $\Delta\nu = 2$. In the figure, the square symbols are obtained from the discharge emission measurements in quiescent air while the diamond symbols represent simulated spectra. In simulating the spectra, we assume that the upper levels in the transition $(C^3\prod_u)$ are distributed in accordance with an assumption of Boltzmann equilibrium with distinctly different temperatures ($T_{\rm rot}$, $T_{\rm vib}$) for the individual rotational and vibrational modes. These temperatures are adjusted in order to obtain the most satisfactory fit between the measured and simulated spectrum, the latter of which is

TABLE I
OBSERVED PROPERTIES OF THREE DISCHARGES INVESTIGATED

	SECD	DBD	USRD
Power source	ac	ac	pulsed
Typical peak voltage	1 kV	5 kV	5 kV
Typical peak current	–	0.5 A	25 A
Typical frequency	–	25 – 40 kHz	15 – 50 kHz
Typical pulse width	–	10 ns (multiple)	10 ns (single)
EM noise	negligible	moderate	very high
Electrode	metal–flame	metal–dielectric	metal–metal
Pulse controllability	impossible	difficult	easy
Degree of nonequilibrium	–	good	better
Power consumption	~ 0.1 W	~ 1 W	~ 10 W
Cost	low	low	high

The symbol '–' represents uninvestigated properties.



(a)

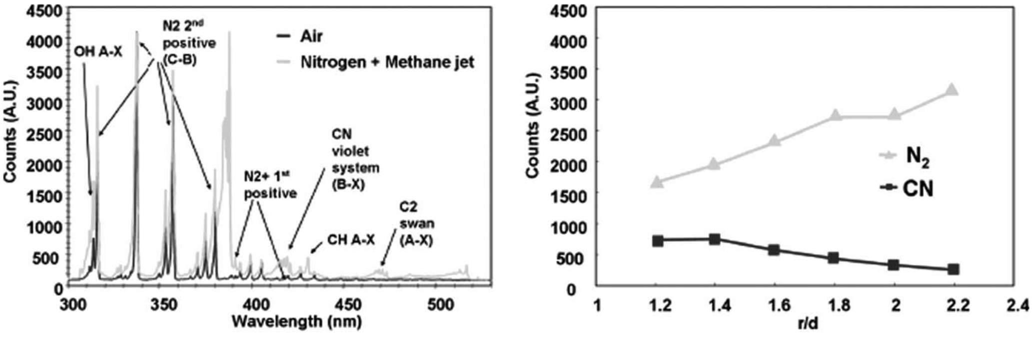
(b)

Fig. 4. (a) Improvement of liftoff jet velocity as a function of normalized coflow speed. Stability limits are extended to $2.5S_\mathrm{L}$, $3S_\mathrm{L}$, and $20S_\mathrm{L}$ with SECD, DBD, and USRD, respectively. The input P-P voltage is 9.35 kV, and the frequency is 30 kHz in SECD and DBD, while USRD uses 6 kV and 15 kHz of frequency. The electrode position is fixed at ten diameters downstream. (b) Sample photographs of a USRD stabilized lifted methane-jet flame. The coflow speed of each case is $5S_\mathrm{L}$ (left), $10S_\mathrm{L}$ (middle), and $20S_\mathrm{L}$ (right).

composed of a line-by-line summation of individual rovibronic contributions, with known line strengths and spectroscopic constants that can be used to compute term energies.

In accordance with the difference in discharge voltage/current profiles discussed above, the resulting spectra are also noticeably different. In particular, the rotational and vibrational temperatures of each discharge indicate a higher degree of nonequilibrium in the USRD case. The higher nonequilibrium temperatures in the USRD has important consequences, particularly in terms of discharge efficiency since the rotational temperature rapidly reaches equilibrium with the translational temperature (which determines the gas heating) while the vibrational temperature is an important parameter that reflects the chemical reactivity of molecules. Thus, we find that the USRD affords not only a higher current density but also potentially higher performance in affecting the flame behavior, largely because of the higher vibrational temperatures and lower rotational temperatures generated in the discharge. In Table I, a comparison of the three discharges is summarized. We believe it is important to note that the typical power consumption of any of the three discharges is less than or equal to 0.1% of the output chemical power of the system.

The dramatic enhancement in flame stabilization due to the application of the nonequilibrium discharges is summarized in Fig. 4(a). Here, the coflow velocity is used as a parameter against which the critical jet velocity for liftoff is compared. For a given normalized coflow velocity $V_\mathrm{CO}/S_\mathrm{L}$, the jet velocity at which the flame blows OFF is plotted, and the stable region is the region under the curves. For this figure, the variation of the jet velocity is achieved by increasing flow rate at fixed nozzle diameter (4.6 mm). The range of the jet Reynolds number is approximately 3000–8000. In comparison to the case without the discharge, where the velocity limit of the pure methane jet varies from 10.5 to 14 m/s ($\mathrm{Re_d} \sim 3000 - 4000$) and the coflow stability boundary is $V_\mathrm{CO}/S_\mathrm{L} \approx 2$, it is shown that the SECD increases the maximum jet velocity by approximately 20%–30% and extends the coflow velocity stability limit by a slight value. The use of the DBD results in a 50%–100% increase in the maximum velocity of the pure methane jet and a modest increase in the coflow velocity limit. It does confirm, however, that the DBD configuration shows more promise in enhancing flame stability than the SECD, most likely due to its ability to support a higher discharge current density (resulting in a higher overall deposited power). A further advantage of the DBD configuration is that the DBD can be sustained in the



(a) (b)

Fig. 5. (a) Emission spectra of various jet mixtures. (b) Emission intensity as a function of discharge position along the radial direction obtained at $x/d = 8.5$ with 6 kV and 15 kHz USRD. The jet Reynolds number is 3991.

absence of the flame, which is in contrast to the SECD configuration, where the flame serves as a virtual ground electrode, and is necessary for sustaining the discharge. It is noteworthy that with the DBD configuration, the maximum coflow velocity limit (where the flame is stably anchored to the electrode) is extended to approximately $3S_{\mathrm{L}}$ even with just a modest discharge power of 0.5 W—only 0.005% of the rate of energy released by the flame itself.

The most significant stability enhancement is seen with the USRD configuration. Use of the USRD (in a pure methane-jet diffusion condition) extends the coflow velocity stability limit by nearly ten-fold. We attribute this to its higher current density (higher dissipated power) than either the DBD or SECD. The power consumption of the USRD in this particular case shown in Fig. 4(a) is approximately 15 W, i.e., considerably greater than either the DBD or the SECD. This higher dissipated power makes a direct comparison (based on an energy budget) between the three different discharge modes difficult. However, it is noteworthy that the ability of the USRD to couple more power into the flame is clearly advantageous.

Finally, the black squares in Fig. 4(a) represent the USRD results for a highly diluted (20% nitrogen, 80% methane) jet mixture to simulate a low-grade fuel. It is apparent that the USRD still results in a significant enhancement of flame stability even in such a highly diluted jet diffusion flame condition. The photographs shown in Fig. 4(b) are examples of methane flames that are stabilized by the USRD. It is apparent that the flame base is unusually distorted due to the high strain rate caused by the high coflow velocity ($\sim 5S_{\mathrm{L}}$, $10S_{\mathrm{L}}$, and $20S_{\mathrm{L}}$).

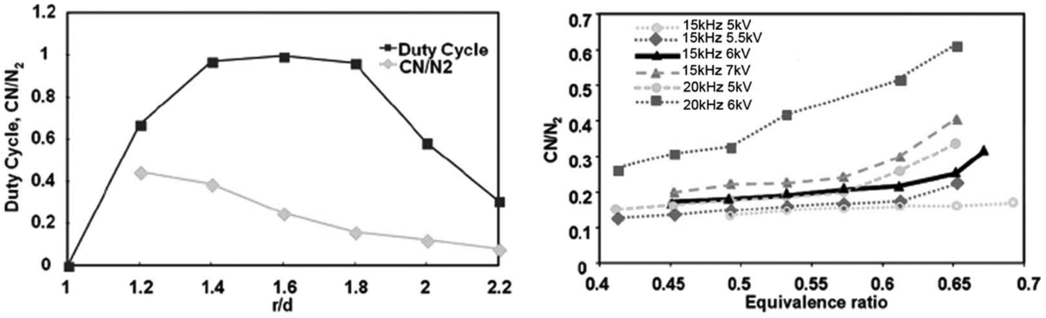
B. *Optimal Discharge Placement*

It is well known that the location where a lifted jet flame is naturally stabilized is closely related to the distribution of a specific fuel/air mixture fraction. For example, Joedicke *et al.* [25] showed that the average position of a naturally stabilized lifted flame base is located on the region of approximately stoichiometric fuel/air mixture fraction ($Z/Z_{\mathrm{st}} = 1.14$).

To investigate the mixture-fraction conditions at optimal discharge placement in physical space, we surveyed physical space by translating the electrode in the jet in coflow configuration while collecting spectral emission data and compared the emission from that of a fully premixed environment of known equivalence ratio. Since the direct measurement of mixture fraction in a turbulent diffusion flame is not trivial, we exploit an advantage of the discharge-emission-spectra change when local mixture fraction varies. Fig. 5(a) compares a typical USRD discharge emission spectrum in a pure air jet to that of a methane/nitrogen jet in a coflow of air. For both the pure air and the methane/nitrogen jet, most of the plasma-excited emission features are attributed to excited molecular nitrogen. However, in the case of the methane/nitrogen jet, we see strong cyano (CN) emission near 388 nm along and CH and C_2 emission near 430 and 450–550 nm, respectively. The plasma excitation spectrum serves to distinguish between methane-containing and nonmethane-containing jets, forming the basis for our quantitative determination of mixture fraction in the diffusion flames.

Fig. 5(b) compares the emission intensity of CN near 388 nm to that of N_2 near 337 nm as a function of jet radial position for various USRD discharge voltages, for the case of a pure methane jet in an air coflow. For this particular case, the jet velocity and normalized coflow velocity are 14 and 1.5 m/s, respectively. This graph indicates that the emission intensities of CN and N_2 are monotonically varying functions of radial position, with the CN signature diminishing as the jet boundary is approached, in contrast to the increased signal from N_2. The ratio of CN to N_2 intensity can be used as an indicator of mixture fraction, when appropriately calibrated. While the relative CN signal provides a measure of methane component, the normalization to N_2 provides a reference that accounts for possible differences in collection discharge volumes and in power dissipation. The intensities are also dependent on discharge peak voltage and frequency requiring these discharge conditions be reproduced in the calibration of the intensity ratios (discussed below) against known mixture-fraction conditions.

Fig. 6(a) illustrates the measured variation in the flame duty cycle with placement of the discharge along the jet radius. For these results, the discharge was at a fixed axial position of $x = 8.5d$, and the discharge voltage and pulse frequency were 6 kV and 15 kHz, respectively. Also shown in the figure is the corresponding measured CN/N_2 emission intensity ratio. It appears that the optimal electrode placement (corresponding to maximum duty cycle) is approximately $1.6d$ outward from the



(a) (b)

Fig. 6. (a) Flame duty cycle coplotted with CN/N_2 ratio. The discharge and flow conditions and electrode location are identical to those in Fig. 5. The optimal position of the discharge corresponds to $CN/N_2 = 0.25$. (b) CN/N_2 ratio in a fully premixed environment. The premixed flow speed is 0.5 m/s. $CN/N_2 = 0.25$ corresponds to 0.65 equivalence ratio under premixed conditions.

center of the jet at this discharge condition and axial location. At this location, the CN/N_2 intensity ratio is found to be 0.25. It is expected that the placement for maximum flame duty cycle will also vary with axial location, however, the CN/N_2 intensity ratio corresponding to any optimal discharge position is unaffected and found to be 0.25.

In order to interpret these findings, a calibration is needed to determine the absolute mixture fraction that corresponds to a value of 0.25 in the CN/N_2 intensity ratio. This calibration is performed with fully premixed flow (using a separate burner) where the CN/N_2 ratio is measured as a function of equivalence ratio for various voltages and pulse repetition frequency. The results of this calibration are shown in Fig. 6(b). The voltage and frequency calibration conditions for the results in Fig. 6(a) are given by the black solid line in the figure. The measured CN/N_2 ratio of 0.25 is seen to correspond to very lean conditions $(Z/Z_{\mathrm{st}} \sim 0.65)$ in contrast to that of a conventional lifted jet flame, where the flame base seems to be at conditions of $(Z/Z_{\mathrm{st}} = 1.14)$, i.e., closer to stoichiometric conditions.

We believe that the mixture fraction at which optimal stability is seen results partly from improved coupling of energy to the flow as the electrode pair is moved OFF axis (note that there is no discharge on axis, or for radial positions $r < d$). At radial positions close to the methane jet/air boundary, it is presumed that the discharge results in energetic electrons and a nonequilibrium energy distribution that leads to electron-induced dissociation and the production of important radicals such as OH, O, and H, which can affect the flame stability. Although not shown here, we find that the emission of the discharge is dominated by electronically excited molecular nitrogen. In addition to direct electron-induced reactions, it is also possible that electronically excited N_2 can channel energy to other species, further enhancing the radical pool. A preliminary reaction path analysis [28] supports the conjecture that energy transfer between excited N_2 and atomic hygrogen (H), atomic oxygen (O), molecular oxygen (O_2), methyl radical (CH_3), and hydroperoxyl radical (HO_2) may lead to increased OH levels, which promotes flame stability. However, further studies are needed to develop a quantitative understanding on the balance between energy coupling and kinetics that leads to an optimum stabilization where the local mixing fraction Z is 65% of the stoichiometric value Z_{st}.

IV. CONCLUSION

Three different types of nonequilibrium discharges were used to enhance the stability of a lifted methane-jet diffusion flame in coflow. An SECD resulted in a marginal stability increase due in part to the low power deposited into the flame. An electrode pair forming a DBD was somewhat more effective than the SECD, extending the coflow velocity limit to over three times the laminar flame speed. The use of an ultrashort repetitively pulsed discharge (USRD) resulted in significant increase in flame stability in part due to higher deposited power in comparison to either the SECD or the DBD. Using the USRD, we have found that a lifted methane-jet flame is stable in coflow velocities of up to 20 times the laminar flame speed.

We found that there is an optimal USRD discharge placement at a fixed axial position for maximum flame stability (highest flame duty cycle ~1). This radial position corresponded to approximately $1.6d$ (at a height of $8.5d$). An analysis of the emission from the plasma and its comparison [through the relative intensity of CN (388 nm) and N_2 (337 nm) bands] to plasma emission generated in a premixed flow of methane and air of known mixture fraction, suggested that the mixture fraction at the optimum discharge placement in the jet diffusion flame corresponded to $Z/Z_{\mathrm{st}} \sim 0.65$, i.e., conditions that are much leaner than those at the flame base of a naturally stabilized flame.

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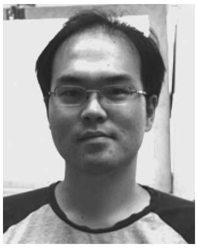

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