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Citation: [AIP Conference Proceedings](#) **1628**, 351 (2014); doi: 10.1063/1.4902614

View online: <http://dx.doi.org/10.1063/1.4902614>

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Dynamic and Hydrodynamic Response of Premixed Methane-air Flame to Nanosecond Pulsed Discharges

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Abstract. Nanosecond pulsed discharges were applied to an experimental study for exploring the temporal response of a premixed methane/air Bunsen flame with two optical diagnostic methods. The microstructure and dynamic response of flame to the discharges was observed by a schlieren system, and the radial velocity of the shock wave was also measured. Time-resolved optical emission spectroscopy offered intermediate species, their evolutionary timescales and temperature in the plasma region. While the ultrafast heating mechanism of NPD was confirmed, the experimental results also provided lots of critical data for understanding the dynamics of non-equilibrium plasma produced by NPD and performing further numerical simulation. In addition, it also indicates that NPD can stir reactant gases and interact with flame by the generated shock wave and vortices while it produces more reactive plasma than LIBS. It makes the technique may be more suited for applications in combustion enhancement and instability control.

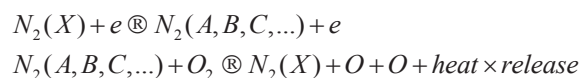
Keywords: Nanosecond pulsed discharges (NPD); schlieren; optical emission spectroscopy (OES); time-resolved
PACS: 52.38.Kd, 52.70.Kz, 47.80.Jk

INTRODUCTION

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Plasma-assisted combustion is a promising way to enhance flame velocity and reduce pollutant emissions[1-3]. Because of the vital practical significance to alleviate the resource and environmental problems, a variety of plasma generator systems including dielectric barrier discharge (DBD) [4], corona discharge[5], and microwave discharge[6], and the nanosecond pulsed discharges[7] have been used to explore plasma-assisted combustion. In these systems, nanosecond pulsed discharges have received special attention due to the high energy efficiency and low power consumption. Furthermore, nanosecond pulsed discharges can produce large amounts of neutral molecules, atoms, radicals, and other active species in mixtures within combustion in nanosecond scale which is shorter than most of chemical reaction time. Therefore, the well-known crucial activated particles can initiate chain reactions, shift chemical equilibrium and accelerate chemical reactions greatly, even at low concentrations. Consequently, nanosecond pulsed discharges technique has been applied to decrease the lean flammability limit[8, 9], increase the laminar flame speed [10, 11], or to reduce the ignition delay time [12, 13] in the past decades. In these investigations, except for studies that aimed to determine the ignition delay, most of researches were made from chemical and thermal activation in stationary conditions, with investigating the influence of various parameters, such as pressure, temperature, fuel, burner geometry, electrode geometry or other plasma properties. The dynamic and hydrodynamic responses of the flame to nanosecond pulsed discharges, which are very important to clarify the theoretical mechanisms or to perform numerical simulation, are largely unknown.

The two-step ultrafast mechanism initially proposed by Popov. [14] is the most popular approach for interpreting electron kinetics in non-equilibrium plasmas induced by NPD. The mechanism is composed of the following two steps:



According to the ultrafast mechanism, nitrogen molecules are firstly excited by electron impact to electronic states such as $A^3\Sigma$, $B^3\Pi$, and $C^3\Pi$ during voltage pulses. Then, excited N_2 molecules are quenched by oxygen molecules, producing atomic oxygen radicals and heat release in the afterglow. In previously studies, the two-step process mechanism was verified by the experimental observation of the ultrafast production of O atom [15] and the

measured high rotational temperature of N_2 with the assumption that the rotational and translational modes of N_2 are equilibrated at all times [16]. More plenty of transient information about the process and direct evidences to further confirm the ultrafast heating are needed in order to prove or supply the mechanism and to understand the non-equilibrium process of NPD.

Therefore, in this paper, we present a fundamental study investigating the transient interaction of a premixed CH_4 /air Bunsen flame with nanosecond pulsed discharges by a nanosecond-gated schlieren system and time-resolved optical emission spectroscopy. The former optical technique was employed to image the shockwave propagation and interaction process of flame and discharges while the latter one was carried out to monitor the active species and the temperature evolution. To our knowledge, both of the two techniques have rarely been used to understand the nanosecond pulsed discharges.

EXPERIMENTAL SETUP

Figure 1 demonstrates the schematic diagram of the experimental setup. In a typical experiment, the high-voltage (HV) signals used for discharging were supplied by a homemade HV alternating current transformer (based on the ET series 2000W Regulated High Voltage DC Power Supplies, Glassman High Voltage Inc.) and monitored by an HV probe (Tektronix P6015A) combined with a digitizing oscilloscope (Tektronix DPO4032). The premixed gas sample containing methane and air with a suitable equivalence ratio, supplied from cylinders through stainless steel pipes and controlled by an individually calibrated mass flow controller (SLA5851S Thermal Mass Flow meter, BROOKS Instrument), flows through distant pipelines and the annular space between the inner electrode and the quartz tube of the Bunsen burner. The mass flow meters controlled the gas flow precisely, while the distant pipelines ensured that gases were mixed sufficiently. The nanosecond pulsed discharges were conducted on the premixed Bunsen burner schematically presented in the insert of Fig. 1. The Bunsen burner is a coaxial configuration composed of an inner copper stick whose diameter is 4mm and an outer quartz tube with an inside diameter of 8mm and the thickness of 1mm. The copper stick was employed as the cathode electrode (grounded) of the NPD. The tungsten anode is a cylindrical electrode which has a diameter of 1 mm with a sharpened tip. In discharges, a positive high voltage (HV) pulse of 5.8 kV was applied to the electrode tip while the inter-electrode gap was set at 2.5 mm. The Full-Width-Half-Maximum (FWHM) duration of the HV pulse is about 90 ns generally. For the measurement of the schlieren photography, a pulsed xenon flash lamp with duration of 800 μs was used. The schlieren set-up consisted of achromatic type lenses with a diameter of 150 mm and a focal length of 1500 mm. The burner was located in the center of the two concave mirrors. Light came from xenon flash lamp and illuminated the laminar premixed CH_4 /air flames. After passing through the knife edge, the schlieren images were recorded by using an ICCD (Model: PI-Max3, minimum optical gate width: 2 ns, Princeton Instruments) with a camera lens (Casiot, $f = 35$ -135mm). The output of the ICCD was input to a digital board and then analyzed using a computer data acquisition system. The flame emission was collected by a lens system, and then, transmitted into an Acton SpectraPro 2300i spectrometer (focal length 300 mm) equipped with an ICCD (PI-Max, minimum optical gate width:

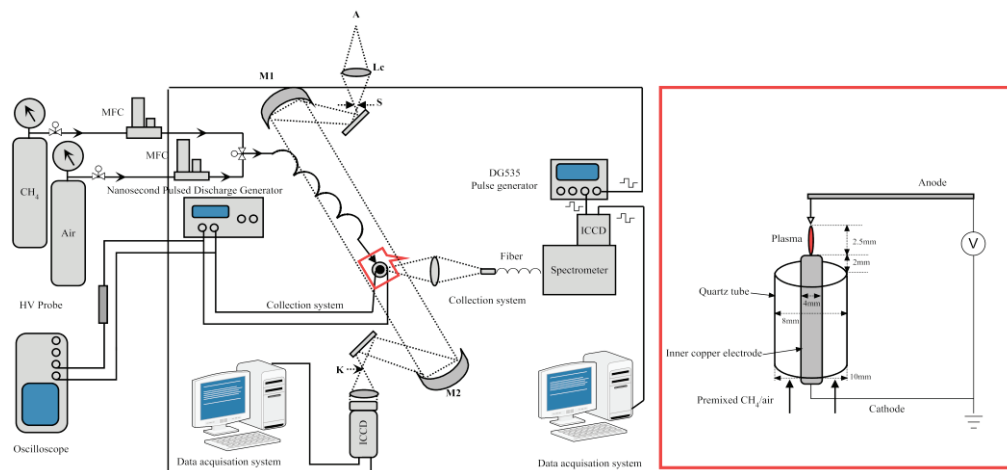


FIGURE 1. Schematic diagram of experimental setup.

2 ns, Princeton Instruments). The spectrometer used is equipped with a 300grooves/mm grating and a 50 μ m-wide entrance slit to get the lower resolution spectra, and with a 1200grooves/mm and 10 μ m-wide slit to get the higher resolution spectra. The grating efficiency of the spectrometer was calibrated using an NIST standard tungsten halogen lamp (Model: 63976, Oriel Instruments) and the wavelength calibration of the system was performed using a standard Hg-Ar lamp. Experimental synchronization was controlled through a multi-channel digital delay pulse generator (Model: DG535, Stanford Research System).

RESULTS AND DISCUSSION

Microstructure Evolution and Hydrodynamic Processes in Plasma Region with Schlieren Photography

Single-shot Successive Schlieren Images of NPD in Air

Schlieren photography technique, a widely used qualitative optical investigative method, is a simple and convenient way to measure the laminar flame speed indirectly, utilizing the deflection of parallel light rays in the presence of variable density gradients normal to the propagation of the ray. In the present experiment, the successive schlieren images of shockwave propagation induced by the single-shot nanosecond pulsed discharges in air were recorded and displayed in Figure 2.

In the figure, it noted that initially the shock-wave accompanies the heated gas channel growth in a cylindrical shape. It means that, during discharges, the NPD leads to a rapid increase in the pressure and temperature in the gas channel forming high-temperature high-pressure plasma channels at first. While the discharge energy is transferred into the excitation of internal degrees of freedom in the gas, a portion of the discharges energy is rapidly converted to the energy of translational degrees of freedom. The fraction of transmitted energy varies from 10% to 60% at different electric field [17]. The process of energy transfer and relaxation occurs in tens or hundreds of nanoseconds, which is less than the typical gas-dynamic time. Then, overheated high-pressure plasma channels begin to expand into the surrounding space forming a strong cylindrical shock wave just like in Fig. 2 (delay=0.1 μ s). Next, the hot, low-density gas of the plasma channel pushes the cold, high-density gas out, causing the propagation of the shock wave and the separation of the shock wave and the heated gas channel. When the shock wave propagates beyond the heated gas channel, it changed from a cylindrical shape to a spherical wave as shown in Fig. 2 (delay=6 μ s). Finally, the spherical-shape shock wave will degenerate into a sound wave, according to the calculated shock wave velocity that will be displayed below. Afterwards, in the large time scale (hundreds of microseconds), the vortexes and turbulization capable of stirring gases in the discharge area appear. It should be induced by the initial overexpansion of the hot gas channel and the reverse flow caused by the electrodes of the discharges.

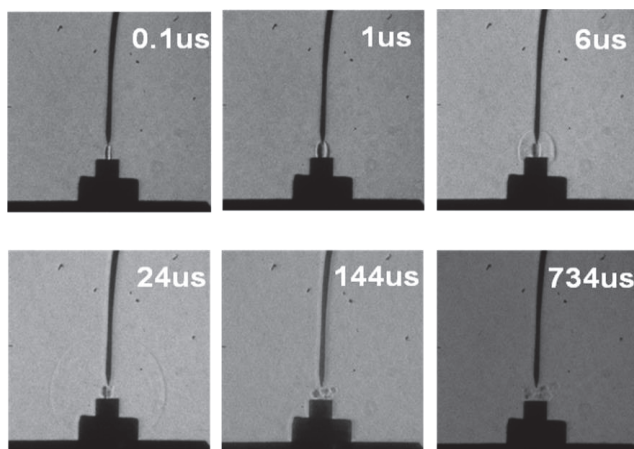


FIGURE 2. Typical schlieren images of single-shot nanosecond pulsed discharges in air. The exposure time is 200 ns. Numbers upper the images represent the acquisition time after discharge initiation (called delay time).

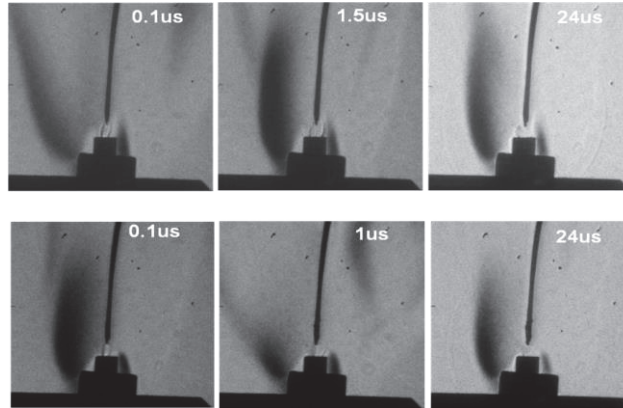


FIGURE 3. Typical schlieren images of single-shot nanosecond pulsed discharges in premixed CH₄/air Bunsen flame (upper row $\Phi=1.27$, lower row $\Phi=0.85$). The exposure time is 200 ns. Numbers upper the images represent the exposure time after discharge initiation.

Schlieren Images of NPD in Bunsen Flame

For the purpose of exploring the interaction of combustion with the NPD, the single-shot nanosecond pulsed discharges were applied to two premixed methane/air Bunsen flames with different equivalence ratio (1.27, 0.85 respectively). Fig. 3 showed that successive schlieren images of shockwave propagation and the interaction process of the single-shot nanosecond pulsed discharges with the flame. Similar to the discharges study in preheated air flow [18], the distinct microstructure of the discharge area was smoothed greatly because of the interaction of flame and the discharges. It is believed that the hot gas flow within the flame reduces the density gradients strongly due to the intense random thermal motion. Moreover, the schlieren images also demonstrated that the reduction in small flame who owns lower equivalence ratio is more obvious. We think that this phenomenon is resulting from the small recirculation area which is deemed to the unburned inner flame in a Bunsen flame. Comparing the radius of the shock wave in flame with that in air at the same delay time, it can be found that the propagation of the shock wave was greatly speeded up. The acceleration will be confirmed and quantized by the measurement of the shock wave speed as shown in the next section.

The Measurement of the Shock Wave Velocity

With the schlieren images of NPD in air and in flame, as shown in Fig. 2 and Fig. 3, the temporal evolutions of the shock-wave radius in the middle plane of the gap are measured. Then, the shock-wave velocity was deduced from the measured radius and the delay time. In Fig. 4, the measured shock-wave velocities in the three mentioned cases were exhibited. From the plots in the figure, it could be found that the shock wave velocities in three cases are all decreased rapidly and exponentially with the delay time and the shock-wave radius.

In air, the radial velocity of the shock wave ranges from about 575 to around 372 m/s during the discharges and its afterglow. Depending on the relation of the temperature and the sound speed, the value of the speed of sound in air at 300 K is 347 m/s that is very close to the measured shock wave velocity after the relaxation process (372 m/s). It means that, as it propagates, the shock-wave induced by discharges degenerates into a sound wave finally. From the difference of the two velocity values, it was inferred that the surrounding air temperature rose by $\sim 70\text{K}$ due to the nanosecond pulsed discharges.

With flame, because of the reduction of combustion, the shock wave was only observed in midway through the full propagation process, as well as the measured velocity. However, it is obvious that the shock wave velocity was significantly enhanced by the combustion. Comparing with the discharges in air, the amplification of the shock wave velocity in flame is quite obvious, even up to 75% at $16\mu\text{s}$ after discharges initiation. At that time, the measured velocities of shock wave in flame are both above 700m/s. It noted that the shock wave velocity and its growth in the small flame ($\Phi=0.85$) are both slightly higher than that in the flame ($\Phi=1.27$). The overlaps of the discharge area and the burned region of Bunsen flame may be the reason.

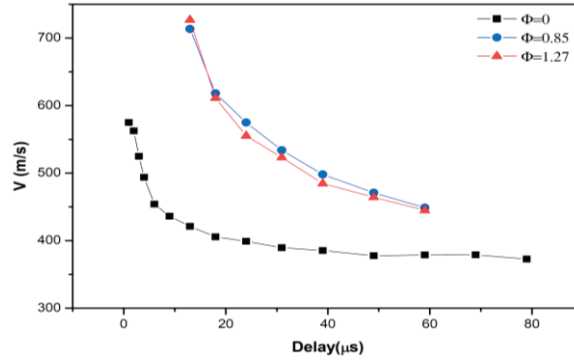


FIGURE 4. The measured radial velocity of shock wave produced by NPD in air and in flame.

As a conclusion, the microstructure and hydrodynamic responses of the flame to nanosecond pulsed discharges were explored by schlieren images. The experimental results demonstrate that the NPD leads to a rapid increase in the pressure and temperature in the gas channel at first, as predicted by the two-step ultrafast mechanism. The overheated high-pressure gas channels begin to expand into the surrounding space forming a strong cylindrical shock wave. Then the shock wave propagates beyond the heated gas channel to constitute a spherical wave. At last, it degenerates into a sound wave. The measurement of shock wave velocity shows that the velocities decrease exponentially with the delay time and the shock-wave radius. And, the Bunsen flame could accelerate the propagation of the shock wave greatly. These experimental results displayed that NPD could generate shock waves and vortices by heating and compressing the gas in the discharge area within a rather short time (short than 100ns in this study). The ultrafast heating and the generated shock wave and vortices may have important applications in aerodynamic flow control, plasma-assisted combustion, or gas treatment.

The Determination of the Transient Species, Their Evolutions and Temperature by Time Resolved Optical Emission Spectroscopy

The Emission Spectra of Bunsen Flame, NPD in Air, and NPD in Flame

In order to determine the intrinsic flame characteristics with NPD and to study the kinetics of the produced active particles capable of affecting combustion, the low resolution emission spectra of three different cases in the range of 300-800nm: case1, the flame optical emission spectroscopy, case2, the spectrum of nanosecond pulsed discharges in air, case3, the spectrum of the flame with NPD, were recorded with millisecond exposure time (500ms, 2ms, 1ms respectively) and displayed in Fig. 5.

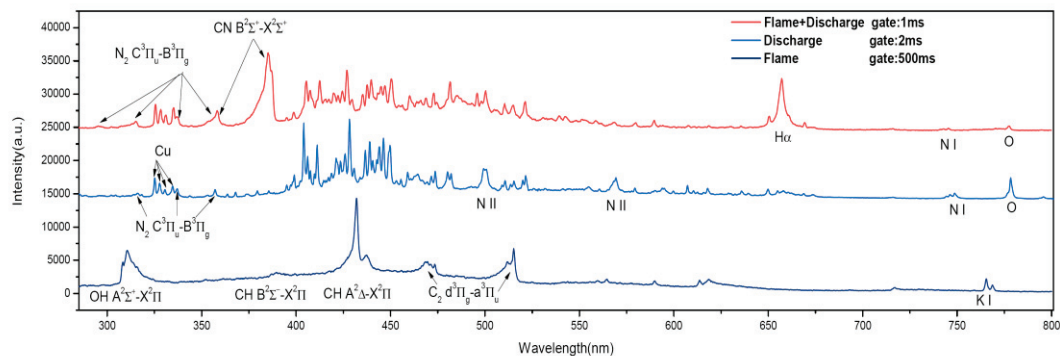


FIGURE 5. Optical emission spectra in three cases: 1, premixed CH_4/air Bunsen flame, 2, NPD in air, 3, NPD in flame ($\Phi=1.27$).

With analyzing the spectra in Fig. 5, it can be found that the emissions of the premixed CH₄/air Bunsen flame are rather weak in case1. Almost all emissions come from the intermediate radicals, e.g. OH, CH, and C₂. In the flame emission spectrum with 500ms exposure time, the A²Σ⁺-X²Π ΔV=0 transition (V is vibrational quantum number) around 308nm of OH radical, the A²Δ-X²Π (~431nm) and B²Σ⁻-X²Π (~ 387nm) of CH radical, and some ro-vibrational bands of the d³Σ_g⁻-a³Π_u electronic transition (the ΔV=0 band head is located at about 516.5nm) of C₂ radical were observed and labeled. It should be noted that all of these weak emissions of flame were not been observed with discharges in a few milliseconds exposure time.

In contrast, the NPD produce large amounts of active atoms and ions whose emissions are quite strong. In case2, according to the NIST Atomic Spectra Database, we had recognized many spectral lines of several atoms and ions, such as the O I triple lines at around 777nm, the lines of N atom (~742.3nm, 744.2nm, 746.8nm), the overlapped lines around 500nm and 569nm of NII in the spectrum of NPD in air with 2ms exposure time. Besides, three extremely weak bands of the C³Π_u-B³Π_g electronic transition of N₂ molecules (located at about 380.5nm, 357.6nm, 337.1nm, respectively) were also determined. In addition, we also find numerous lines of Cu atoms located at 324.7nm, 327.4nm 330.8nm and 333.8nm respectively. It is believed that the active Cu atoms are originated from the copper cathode electrode during discharges.

In case 3, the discharges were carried out within the Bunsen flame. Comparing with that in case2, there are three major changes happens in the spectrum of case3. Firstly, two new species appear. One is the H atoms which came from the fuel (CH₄) in combustion, and the other one is the CN radicals which might be produced by the dissociation and the recombination process during the interaction of combustion and discharges. Secondly, emissions of N₂ molecules were enhanced greatly. It means that the excitation of N₂ molecules is easier in combustion than in air by NPD. Thirdly, the emissions of N and O atoms were reduced while the spectral lines of N ions vanished. It shows that the ions and atoms were involved in the chemical reactions within the flame during the discharges.

The Time Resolved Emission Spectra of NPD in Flame

As mentioned above, these results of emission spectra not only displayed that the interaction of the flame and the discharges, but also offered numbers of experimental evidences for modeling theoretical mechanisms and performing numerical simulation, such as the intermediate species: N₂ molecules, O, N, H atoms, CN radicals and the N ions. However, for modeling or verifying the dynamic mechanism of the nanosecond pulsed discharges, the time scales of the generation, evolvment and disappearance of these species might be more useful. Therefore, the time-resolved emission spectra with nanosecond exposure time were recorded, analyzed, and exhibited in Fig. 6, for determining the transient species and temperature in the inter-electrode region produced by nanosecond pulsed discharges.

These time-resolved emission spectra of the flame with NPD were obtained with the 300grooves/mm grating and a 50μm-wide entrance slit in the wavelength range of 290-400nm. The equivalence ratio of premixed CH₄/air gas was set at 1.27. The gate width of ICCD was programmed as 100 ns while the time of data acquisition varied from 70ns before the discharges initiation to 1930ns after it (delay=-70 - 1930ns). In the time-resolved emission spectra shown in Fig. 6, we had identified the B²Σ⁺-X²Σ⁺ ΔV=0 transition (its head is located at about 388.6nm) and ΔV=-1 transition (~ 359.2nm) of CN radicals (the transitions are also called as the CN violet band), and a series of ro-vibrational spectroscopy, like some bands in ΔV = -2, -1, 0, +1, +2 transitions of the C³Π_u-B³Π_g electronic transition of N₂ molecules, located at about 380.5nm, 357.6nm, 337.1nm, 316.4nm and 298.5nm respectively. In addition, the spectral lines of Cu atoms referred to above were also ascertained.

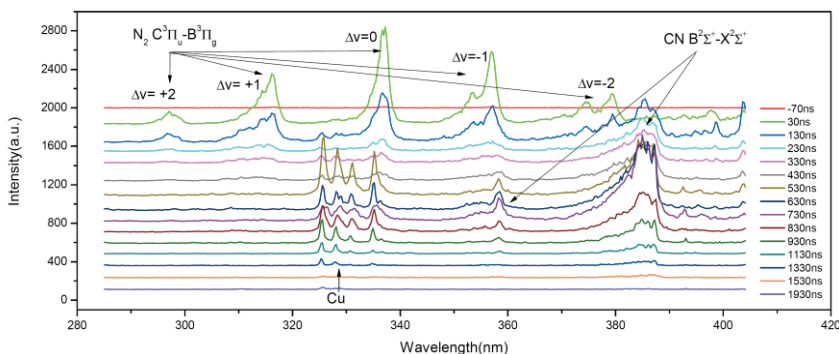


FIGURE 6. Time-resolved emission spectra of Premixed CH₄/air Bunsen flame with NPD.

In Fig. 6, it can be readily found that the emissions of different species appear in an orderly manner. As the discharges initiation, the emissions of N_2 molecules first appeared in several ten-nanoseconds which is considered as the timescale of electron impact during the discharges pulse. And then, the emission of N_2 molecules disappeared in a very short period of time (about 300 ns in this experiment) with the rising of the CN violet band. Comparing with emissions of N_2 molecules, the rising of CN violet band emission is rather slow. It took about 700ns for the climbing while its declination also spent about 700ns. The timescale of the evolution is consistent with that of V–T energy transfer and chemical reactions in the dynamics of NPD. Besides, just like the CN violet band, the spectral lines of H and O atoms ($H\alpha$ at around 656nm and O I triple lines at 777nm, not shown here) also appear at about 100ns after the discharges initiation with the declination of N_2 molecules emissions. The effective lifetimes of them (104ns and 113ns respectively) are distinctly shorter than that in the same flame measured with laser induced breakdown spectroscopy (LIBS), where the lifetimes of $H\alpha$ and O I triple lines are both over than 250ns. We attribute the shortening to the high density of species in the discharge area produced by NPD. Therefore, it can be inferred that the plasma induced by NPD is of higher particle density and more reactive than that in LIBS.

Moreover, the transient evolution of the intermediate species shown by the time-resolved emission spectra in Fig. 6 confirms the ultrafast heating mechanism of nanosecond pulsed discharges proposed by Popov^[14]. As referred in the introduction, the mechanism declared that during the high voltage pulse, nitrogen molecules will first be excited by electron impact to electronic states such as $A^3\Sigma$, $B^3\Pi$, and $C^3\Pi$. Then, excited N_2 molecules will be quenched by oxygen molecules producing atomic oxygen radicals and ground-state N_2 molecules within a few tens of nanoseconds. In the present experiment, the first step of the mechanism was proved by the observed short-life emissions which are originated from the $C^3\Pi_u-B^3\Pi_g$ electronic transition of N_2 molecules in the initiation of discharges, as shown in Fig. 6. Furthermore, the second step also could be verified by the disappearance of N_2 emission and the emergence of the O I triple spectral lines at the time around 100ns after the discharges initiation.

Furthermore, the spectral lines of N atoms and N ions observed in the emission spectra of NPD indicate that the nitrogen molecules were not only excited as declared in the ultrafast heating mechanism, but also were dissociated and ionized by the discharges. The violet bands of CN radicals further displayed that the N_2 molecules, N atoms or N ions were involved in the reactions with CH_4 . Nevertheless, these indispensable intermediate species and the related reactions in NPD were not found in the existing kinetic simulations of the nanosecond pulsed discharges for methane/air combustion, such as GRI-Mech mechanism [19], 2-D kinetic simulations [20].

The determination of temperature of CN radicals

In order to provide more experimental data for further numerical simulation, an accurate measurement of rotational and vibrational temperatures using CN radicals was also preformed in the present investigation. As shown in Fig. 7a, the rotational and vibrational temperatures of CN radicals were measured by simulating the experimental spectra with LIFBASE software, with considering the collisional broadening and Doppler broadening in Bunsen flame. The simulated temperatures and their time evolution with the delay time were displayed in Fig. 7b. For clarity, the delay time in the figure was set as the delay of the acquisition plus the half of ICCD exposure time.

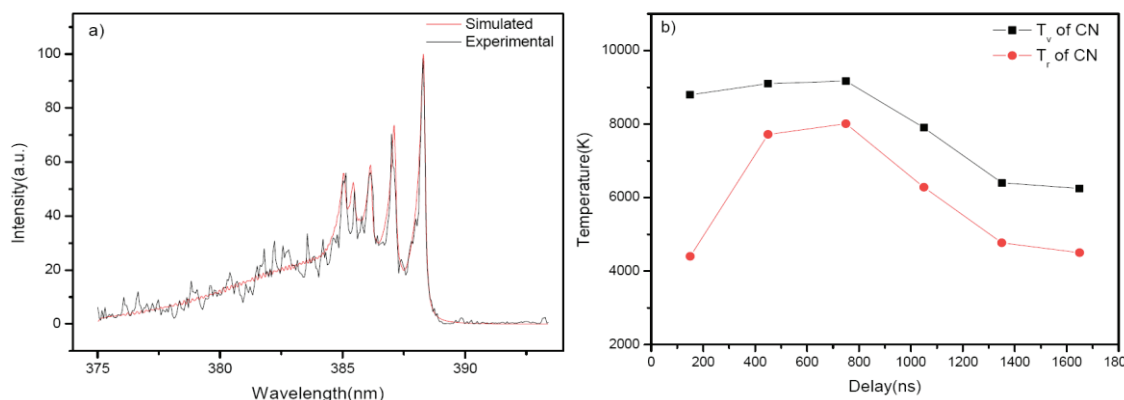


FIGURE 7. a) Experimental and simulated CN emission spectra ($T_v=7900k$, $T_r=6280k$, delay=1050ns, gate=300ns). b) The time evolution of simulated vibrational and rotational temperatures of CN radicals (the delay time in figure was set as the delay of acquisition plus the half -width of ICCD gate).

Simulated temperatures of CN radicals exhibited in Fig. 7b show that, in the rising process of CN violet band emission (delay: 0-700ns, deduced from Fig. 6), the vibrational temperatures of CN radicals are rather high and fairly constant (around 9 thousand Kelvins) while the rotational temperatures grow up from 4400K to about 8000K. It manifests that the formation of CN radicals is an endothermic process with high initial vibrational energy in non-equilibrium plasma. During the declination of CN violet band emission (delay: 700-1500ns), the vibrational and rotational temperatures are both decrease. It indicates that the processes of energy transference and relaxation of CN radicals happen in this period.

Since the collisional rotational-translational (R-T) relaxation occurs within a nanosecond at this high pressure (1 atm), we can expect that gas temperature follows the rotational temperature very closely. And that, gas temperatures affect the physical and chemical processes as well as the energy distribution and equilibrium condition exhibited in the plasma region. Therefore, the measurement of temperatures of CN radicals which is a signature of the chemical reaction between CH₄ and N₂ within the interaction of Bunsen flame and the NPD, especially of the rotational temperature, owns special significance for the heating load estimates related to discharges, understanding the dynamics of non-equilibrium plasma produced by NPD, and performing numerical simulation.

CONCLUSIONS

The transient processes associated with the interaction of a Bunsen flame and the Nanosecond pulsed discharges (NPD) were explored experimentally with two optical methods. A nanosecond-gated schlieren system was employed to explore the microstructure and the hydrodynamic response of flame to the discharges. In the meantime, time-resolved Optical Emission Spectroscopy (OES) measurements were carried out to determine the transient species and temperature in the plasma region created by the nanosecond pulsed discharges. Consequently, the unsteady process of the interaction of flame with the discharges was recorded in real-time by the combined measurements. Numbers of experimental evidences for understanding the dynamics of non-equilibrium plasma produced by NPD and performing further numerical simulation were offered by the experimental results while the two-step ultrafast mechanism was confirmed.

ACKNOWLEDGMENTS

The work is partially supported by National Science Foundation of China (Grant No.11102215 and Grant No. 91216101). The authors acknowledge the technical assistance provided by S.Z. Zhang at the Institute of Mechanics, Chinese Academy of Sciences.

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