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We first investigate the effects of femtosecond-laser-induced plasma on the flame speed of a laminar premixed methane/oxygen/nitrogen flame with a wide range of the equivalence ratios (0.8–1.05) at atmospheric pressure. It is experimentally found that the flame speed increases 20.5% at equivalence ratios 1.05. The self-emission spectra from the flame and the plasma are studied and an efficient production of active radicals under the action of femtosecond (fs)-laser pulses has been observed. Based on the experimental data obtained, the presence of oxygen atom and hydrocarbon radicals is suggested to be a key factor enhancing flame speed. © *2010 American Institute of Physics*. [doi:10.1063/1.3457384]

Recently, increasing the efficiency of fuel combustion and decreasing of pollutant emissions in aircraft engines, gas turbines, and internal combustion engines and so on have become important scientific and engineering problems. To solve these problems, it is necessary to lower barrier of the chemical reaction, enhance burning rate, and expand flammability limits toward lean mixtures. Some basic research is currently performed to study the additional energy effects on reacting flows; one aspect of such research is the investigation of the influence of plasmas on combustion of gaseous hydrocarbons. Various experimental works have reported that atmospheric pressure plasma discharges, i.e., single-electrode corona, dielectric-barrier discharge (DBD), and repetitive ultrashort-pulsed discharge, can enhance flame stability, flame speed, and combustion chemistry.[1–4] However, the plasma discharge electrodes which cannot be changed or transferred after being installed are installed on inner wall of the combustor, so augmented combustion reactions must be restricted in near-wall regions. These problems could be solved by the use of laser-induced plasma because it has many advantages, i.e., controllability of input energy and its duration, and their nonintrusive nature. Moreover, with the absence of electrodes, heterogeneous effects and wall heat loss can be eliminated and it can induce highly reactive plasmas wherever and whenever needed because the laser beam can be focused at any three-dimensional spatial points wherever accessible.

It has been known for a long time that laser-induced gas breakdown process can be used to ignite gaseous combustible mixtures.[5–7] However, there are much fewer studies to date on laser-induced plasma to enhance flame speed and improve flame stabilization which depend on the energy deposition and the lateral growth of the deposited volume. Such a demand makes a single pulse deposition for this application highly unlikely because the lateral expansion of a laser spark might not be strong enough to span across the flow field. Periodic energy deposition might be of practical importance because the deposited pulse energy needs to be sufficient only to initiate a self-sustaining outward flame propagation which eventually interacts with the flames from the preceding and subsequent pulses. Thus, in the repetitive case, a discrete stream of propagating spherical flame is generated. As a result, the burned gas region with its flame front expands laterally.[5] So, to enhance flame speed and stabilize the flame, the plasma must be sustained for a relatively long duration, which requires the laser pulses with high frequency and great irradiance ($\geq 10^{10}$ W/cm^2).[8–10] The femtosecond (fs) laser can generate a train pulses with a maximum irradiance of about 10^{14} W/cm^2 at a frequency of up to 1 kHz in our study. In this paper, we detail the effects of fs-laser-induced plasma (FLIP) on the burning rate of the laminar premixed methane/oxygen/nitrogen flame by examining flame appearance, flame speed, and analyzing the radical species of flame with and without plasma enhancement. For this purpose, we employed two diagnostic tools including two-dimensional flame imaging and time-averaged emission spectroscopy.

Figure 1 is a schematic of the experiment scheme of the plasma-assisted combustion (PAC) in our experiment. For producing plasma, fs-laser pulses of 2 mJ during 40 fs with 800 nm wavelength at a frequency of 1 kHz from a regenerative amplifier were used. The exciting laser beam was focused with lens L1 ($f=25$ cm) into a Bunsen burner along the burner axis. The focal spot size was estimated to be about 44 μm in radius, giving a maximum peak intensity of about 10^{14} W/cm^2. The focal spot was at a distance of about 3 mm to the exit of the burner downstream of mixture. The Bunsen burner that is a straight quartz tube with inner diameter (D) 7.6 mm and length 450 mm is employed. The burner diameter and length are chosen to ensure that the flow remains laminar (Reynolds number, $\mathrm{Re}_D < 2300$) and the flame prevents flashback and blowoff. For performing time-averaged emission spectroscopy, the self-emission from the flame and the plasma was collected and imaged onto an ANDOR SR-

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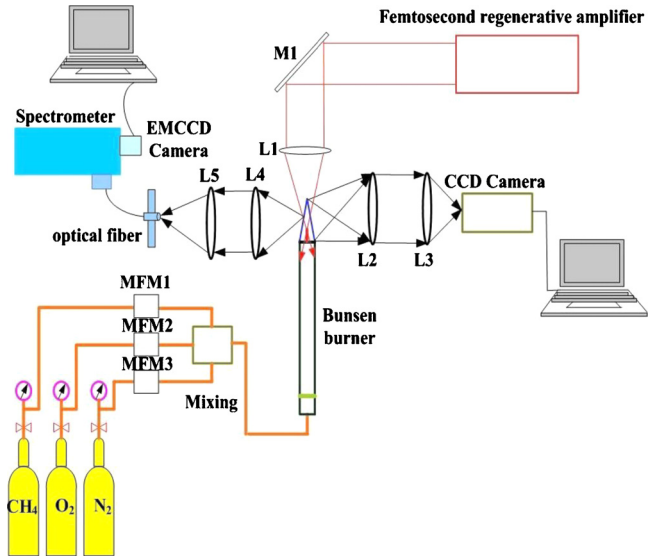

FIG. 1. (Color online) Schematic of the experiment setup (MFM).

750-B1 spectrometer (focal length 750 mm) fitted with a 1600×200 pixel electron-multiplying CCD (EMCCD) camera (ANDOR DU420A-BU2). The resolution (full-width at half-maximum) of the spectrometer is 0.026 nm in the range 200–1100 nm.

For investigating the effect of FLIP on the laminar premixed methane/oxygen/nitrogen flame speed, we utilized the Bunsen flame approach[11,12] to measure the flame speed. The flame speed can be shown as

$$S_u = \frac{\dot{m}}{\rho_u A_b} = \frac{\dot{Q}}{A_b}, \tag{1}$$

where ρ_u is the density of the unburned mixture, S_u is the flame speed of the laminar premixed methane/oxygen/nitrogen flame, A_b is the surface area at the end of the heat release zone and $\dot{Q}$ is the volumetric flow rate of the unburned mixture. Since chemiluminescence is primarily produced in the thin heat release zone of the flame, the surface area measured from a chemiluminescence image can approximate A_b. The volumetric flow rates of CH_4, O_2, and N_2 are recorded separately by thermal mass flow meters (MFM).

Digital images of the flame emission are captured with a 12-bit scientific charge-coupled device camera (1024×1032 pixels) and a telescope system. The camera system is of high sensitivity in the ultraviolet and visible regions ($\sim$300–700 nm), large dynamic range and large well capacity. Some typical images of the flame radiation are given in Fig. 2. As shown in Fig. 2, the plasma changes the flame shape and reduces the reaction zone area. From Eq. (1), while the volumetric flow rate of the unburned mixture is constant and the reaction zone area is reduced, the flame speed must be increased.

In this paper, the equivalence ratio (Φ) is defined as

$$\Phi = \frac{CH_4/O_2}{(CH_4/O_2)_{st}}, \tag{2}$$

where $(CH_4/O_2)_{st}$ refers to the stoichiometric value of (CH_4/O_2). Figure 3 shows the measured flame speeds (S_u) of laminar premixed $CH_4/O_2/N_2$ flame with and without FLIP, the trend of flame speeds versus equivalence ratio are the same. The relevant uncertainties of the measured flame

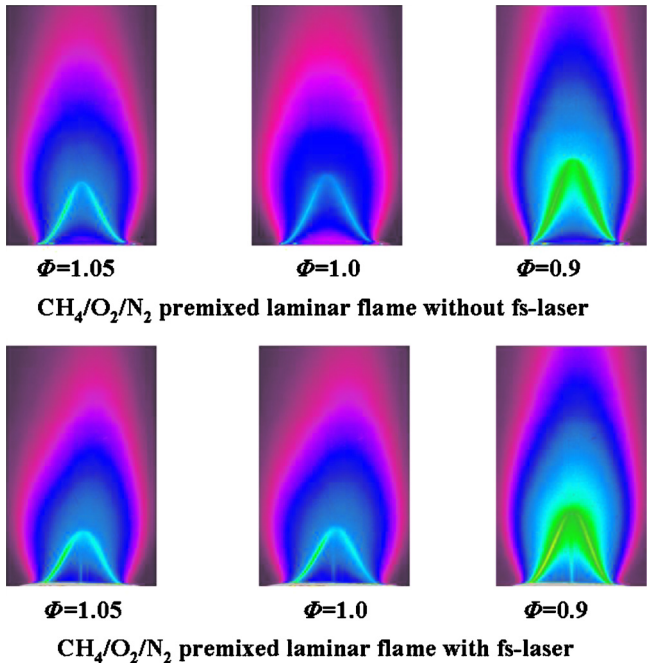

FIG. 2. (Color online) Images of flame for various equivalence ratios (Φ).

speeds of laminar premixed flame with and without FLIP are less than 2.5% and 2.0 %. The FLIP markedly enhances the flame speed which is increased 5.3%–20.5% as the equivalence ratio varies from 0.8 to 1.05. When the equivalence ratio is under 0.9, the effects of the FLIP are little and the laminar flame speeds increases slightly. This is mainly because the CH_4 is lean and the probability that the active atoms and molecules generated by the FLIP react with CH_4 is small.

To determine the intrinsic flame characteristics and to study the kinetics of the active particles capable of affecting the flame speed, we report a detailed comparison between flame radical emission spectra with and without FLIP. Figure 4 shows the emission spectrum of laminar premixed $CH_4/O_2/N_2$ flame with and without FLIP. In the flame emission spectrum, there are the (0, 0) band of A $^2\Sigma^+$–X $^2\Pi$ electronic transition of OH radical ($\lambda=308.9$ nm), and the (0, 0) band of A $^2\Delta$–X $^2\Pi$ electronic transition of CH radical ($\lambda=431.4$ nm), as well as the (0, 0) band of A $^3\Pi_g$–X′ $^3\Pi_u$

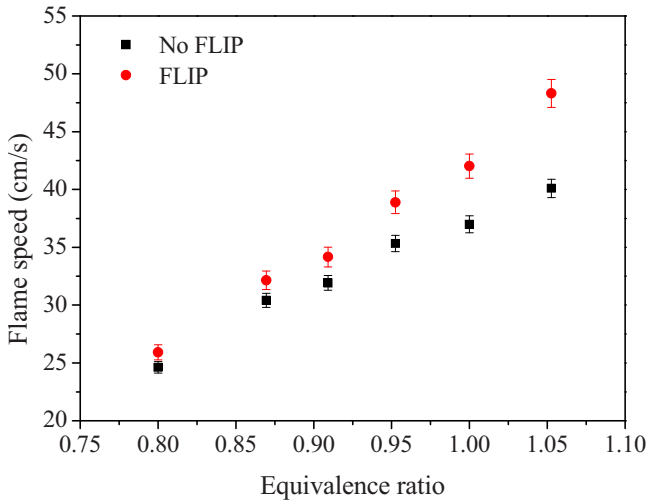

FIG. 3. (Color online) Flame speeds of laminar premixed $CH_4/O_2/N_2$ flame ($38:62O_2:N_2$) with and without FLIP at $p=1$ atm and $T_u\approx300$ K.

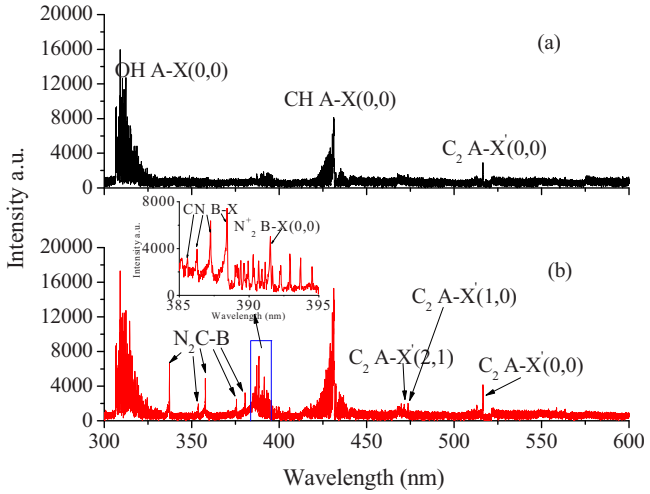

FIG. 4. (Color online) Emission spectra: (a) laminar premixed $CH_4/O_2/N_2$ flame without FLIP; (b) laminar premixed $CH_4/O_2/N_2$ flame with FLIP.

electronic transition of C_2 radical (λ=516.5 nm). In the presence of plasma, besides molecular bands mentioned above, the spectrum contains the (0, 0),(1, 1), (2, 2), and (3, 3) bands of B $^2\Sigma$–X $^2\Sigma$ electronic transition of CN radical (λ=388.3, 387.1, 386.2, and 385.5 nm), and the (1, 0) and (2, 1) bands of A $^3\Pi_g$–X′ $^3\Pi_u$ electronic transition of C_2 radical (λ=473.7 and 471.5 nm). The spontaneous emission of OH increases by at least 10%, and the spontaneous emission of CH and C_2 radicals increase by at least 50% at their peak (Fig. 4). OH, CH, CN, and C_2 radicals play important roles in combustion and are major factors determining both the reaction rate and the flame speed.

To determine the chemical nature of FLIP process, we recorded spectra of FLIP in N_2/O_2 current and laminar premixed $CH_4/O_2/N_2$ flame to measure the species generated just after the laser pulse. As shown in Fig. 5, the oxygen atom at 777.2, 777.5, 777.6和 and 844.8 nm appear in the spectra of FLIP in N_2/O_2 current, and the absence of these emission features in flame indicates that the oxygen atoms are consumed or quenched. Oxygen atom is key specie for the initiation of combustion as it is the main specie responsible for breaking C–H bonds in hydrocarbon

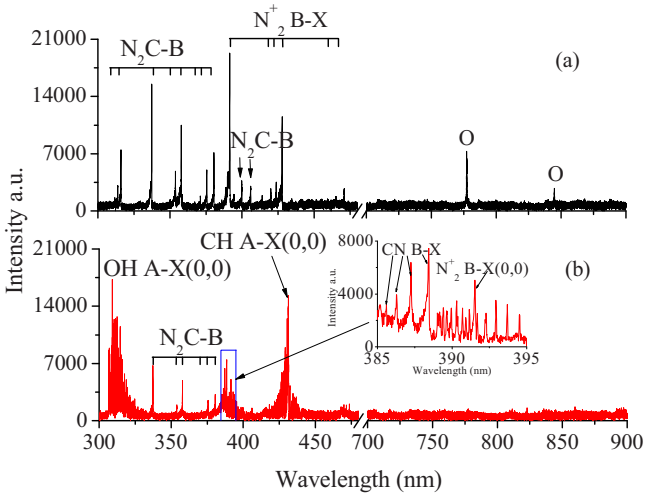

FIG. 5. (Color online) Emission spectra: (a) FLIP in N_2/O_2 current; (b) FLIP in laminar premixed $CH_4/O_2/N_2$ flame.

fuels.[13,14] It also accelerates the chain-branching reactions of methane combustion.

The molecular band of the second positive system of N_2 (the transition C $^3\Pi_u$–B $^3\Pi_g$) in the range 300–480 nm is well pronounced and the (0, 0), (0, 1), (0, 2), (1, 2), (1, 3), and (2, 3) bands of the first negative system of the N_2^+ ion at 391.4, 427.8, 470.9, 423.7, 465.2, and 419.9 nm, respectively, appear in the spectra of FLIP in N_2/O_2 current and at a reduced level in the spectra of the plasma in laminar premixed $CH_4/O_2/N_2$ flame (Fig. 5). These excited N_2 and N_2^+ appear to be consumed or quenched in the flame. Previous studies[13,14] have shown the reactivity of metastable $N_2(A\ ^3\Sigma_u^+)$ with CH_4 to produce $CH_{n,n<4}$ radicals. And the electronically excited nitrogen molecules are quenched by oxygen molecule to produce oxygen atom[1,15] which mainly proceeds through the following reactions:

$$N_2(A\ ^3\Sigma_u^+) + O_2 \rightarrow N_2(X\ ^1\Sigma_g^+) + O + O, \quad (3)$$

$$N_2(B\ ^3\Pi_g) + O_2 \rightarrow N_2(X\ ^1\Sigma_g^+) + O + O, \quad (4)$$

$$N_2(C\ ^3\Pi_u) + O_2 \rightarrow N_2(X\ ^1\Sigma_g^+) + O + O. \quad (5)$$

In conclusion, we demonstrate the possibility to enhance combustion efficiency of a laminar premixed flame at lower power through the use of fs laser-induced plasma. The plasma markedly enhances the laminar flame speed (up to 20.5%). Through spectroscopic measurements, we have evidenced increase in the concentration of free radicals (OH, CH, C_2, and CN) in the flame that may explain the observed enhancement in combustion. It is believed that hydrocarbon radicals and oxygen atom are key species in driving favorable chemistry for combustion enhancement.

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