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Brief Communications

Plasma assisted ammonia combustion: Simultaneous NO_x reduction and flame enhancement

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ABSTRACT

This work reports that plasma can simultaneously reduce NO_x emission and extend the lean blowoff limits of ammonia flames. The results show dramatic differences from similar work using hydrocarbon fuels in which plasma promoted NO_x emission. Therefore, this work could lay the foundation of plasma application in practical ammonia combustion.

1. Introduction

Ammonia combustion features high NO_x emission and poor stability [1]. It is well known that plasma can enhance hydrocarbon combustion and flame stability; however, NO_x formation is promoted simultaneously [2]. There are no experimental studies on plasma-assisted ammonia combustion in the literature to the authors' knowledge. The only relevant works are two numerical studies showing increased flame speeds and reduced ignition delays of NH_3 with electric field/plasma activation [3,4]. Therefore, it is of great interest and importance to investigate the effect of plasma on ammonia combustion. In this work, we focus on NO_x emission and flame stability.

2. Experimental setup

Figure 1(a) depicts the experimental setup for the premixed swirling flame employed in this study. Details of the setup are described in Ref. [2]. Non-equilibrium plasma is generated by a nanosecond high-voltage pulse generator (FPG 50–50MC2 from FID) between the center copper electrode (diameter of 5 mm) and outer ring (diameter of 19 mm) at the nozzle exit. Images in Fig. 1(b) to (e) show direct photographs of ammonia/air flames without and with plasma (discharge voltage V = 11 kV, pulse repetition frequency f = 7 kHz, 39 W). The ranges of voltage and frequency used in this study are 6 to15 kV and 2 to 26 kHz, respectively.

NH_2^* chemiluminescence is measured near 630 nm (α-band) [5–7] using a bandpass filter centered at 632 nm with ±5 nm full width at half maximum. Images are captured using a digital DSLR camera (Sony A55) or a CMOS camera (NAC GX-3) equipped with an intensifier (Ultracam3). Exhaust gas is sampled at the center of the quartz tube exit and measured by a gas analyzer (Horiba PG-350) for NO_x (combined NO and NO_2) and O_2 concentrations. There is no noticeable back diffusion effect near the tube exit, and the composition of exhaust is uniform along the radial direction, which was demonstrated by monitoring O_2 concentrations at different locations near the tube exit.

3. Results and discussion

Figure 2(a) shows the lean blowoff limits (LBO) of ammonia/air flames with and without plasma. Without plasma, an initially attached, stable flame (Fig. 1(b)) is lifted (Fig. 1(d)) from the dump plane with the increase of air flow rate (fuel flow rate is fixed). If the equivalence ratio (ϕ) is decreased below the values indicated by the black line in Fig. 2(a), blowoff occurs.

With plasma activation, flames are always stabilized near the burner exit (Fig. 1(c) and (e)). With plasma at V = 11 kV, f = 7 kHz (discharge power 39 W, corresponding to 1.9% of thermal load at ϕ = 0.94), LBO is significantly extended as shown by the red dashed line in Fig. 2(a). With constant discharge power of 39 W, if V is increased to 15 kV (f = 4 kHz), LBO is further extended as indicated by the blue dashed line in Fig. 2(a). However, if V is decreased to 6 kV (f = 26 kHz), the effect of the plasma becomes negligible. While V decreases from 11 kV to 6 kV, the discharge transits from filamentary discharge to corona discharge, which is more localized and only exists near the rim of the electrodes. Since the discharge power is constant, the observed sensitivity of LBO to discharge

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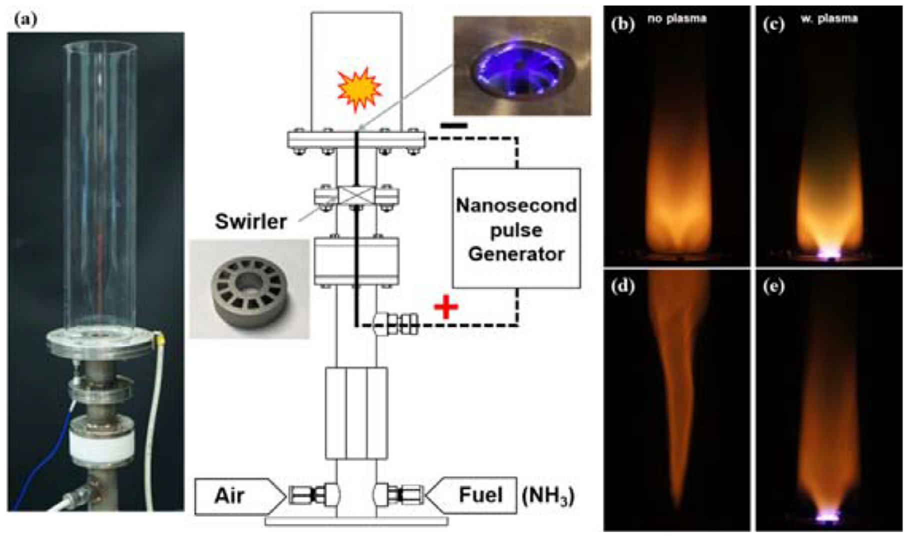

Fig. 1. (a) Schematic of the experimental setup, and direct photographs (ISO100, F/10, 2 s) of flames (b) no plasma, ϕ=0.94 (c) with plasma, ϕ=0.94, (d) no plasma, ϕ=0.71 (c) with plasma, ϕ=0.71.

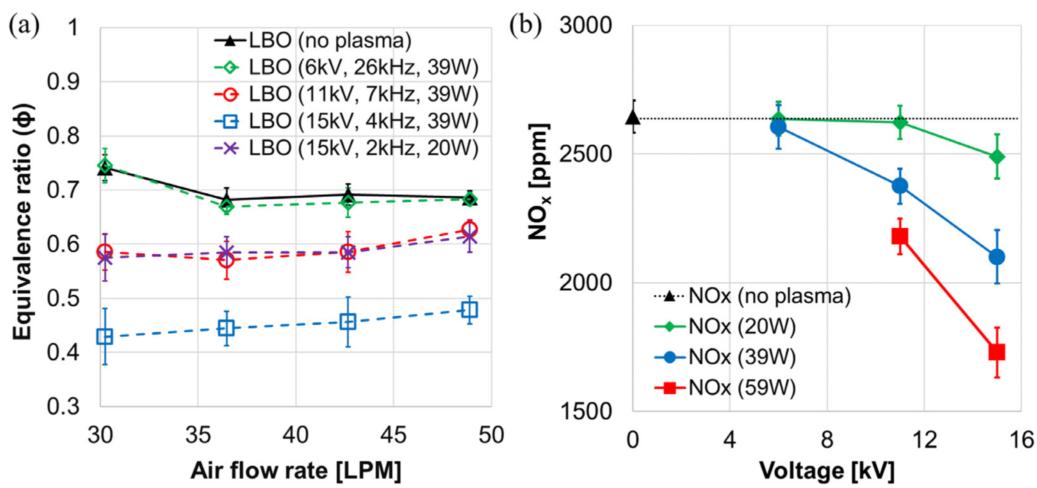

Fig. 2. (a) lean blowoff limits of ammonia/air flames with and without plasma, (b) NO_x emissions without and with plasma (ϕ = 0.94).

voltage (reduced electric field) indicates a kinetic effect in this instance. However, if V is kept constant at 15 kV and f is decreased to 2 kHz (corresponding to 20 W discharge power), the extension of LBO decreases becoming comparable to the case with 39 W discharge power and 11 kV. The discharge power is proportional to the increase of both reactant temperature and number of reactive species produced by the plasma. However, 39 W thermal power only increases the average reactant temperature by 6 K, which has very limited effect on LBO. Therefore, the effect of plasma on LBO might be primarily kinetic in nature. Detailed diagnostics are thus warranted to further understand the underlying physics.

Measured NO_x concentrations are shown in Fig. 2(b). Without plasma, ammonia/air combustion produces approximately 2645 ppm NO_x at ϕ=0.94 (calculated adiabatic flame temperature is 2007 K). With plasma, NO_x emissions are significantly reduced. The corresponding adiabatic flame temperature is 2009 K, assuming 59 W plasma power is purely thermal in nature. It is also surprising to see that NO_x concentration further decreases with the increase of both discharge power and voltage. NO_x concentration decreases as the discharge voltage of plasma is increased at constant discharge power. If the discharge voltage is fixed, NO_x concentration decreases with the increase of discharge power (via increased f). The tendency of NO_x formation is opposite to that for methane/air flames [2]. Recent work from Zhong et al. [8] on plasma assisted n-dodecane/O_2/N_2 pyrolysis and oxidation showed that NO formation was reduced owing to formation of the $C_{12}H_{25}O_2$ radical which consumes NO at low temperature conditions. Zhong et al. [8] also reported that higher discharge frequency and higher plasma discharge power resulted in more NO reduction. In the case of NH_3 oxidation, it is possible that a large quantity of HO_2 is formed in the plasma region and that this HO_2 consumes NO and NO_2 through reactions $NO+HO_2 \rightarrow OH+NO_2$ and $NO_2+HO_2 \rightarrow HONO+O_2$.

Another possible reaction pathway to consume NO is through reactions with NH_2: $NO+NH_2 \rightarrow NNH+OH$ and $NH_2+NO \rightarrow N_2+H_2O$, both of which are essential in the thermal De-NO_x process [9]. Pugh et al. [5] showed a reduction of NO concentration with an increase in NH_2^* chemiluminescence when the equivalence ratio increased. NH_2^* chemiluminescence may be used as a possible indicator of NH_2 production [10] and NO reduction.

Figure 3 shows instantaneous ((a) and (b)) and averaged ((c), (d), (e) and (f)) NH_2^* chemiluminescence, acquired with the intensified CMOS camera. With plasma, NH_2^* chemiluminescence (Fig. 3(b)) is significantly brighter and shows a clear "V" shape (inner shear layer) compared to that without plasma (Fig. 3(a)), indicating additional NH_2^* is generated by the plasma. If ϕ is de-

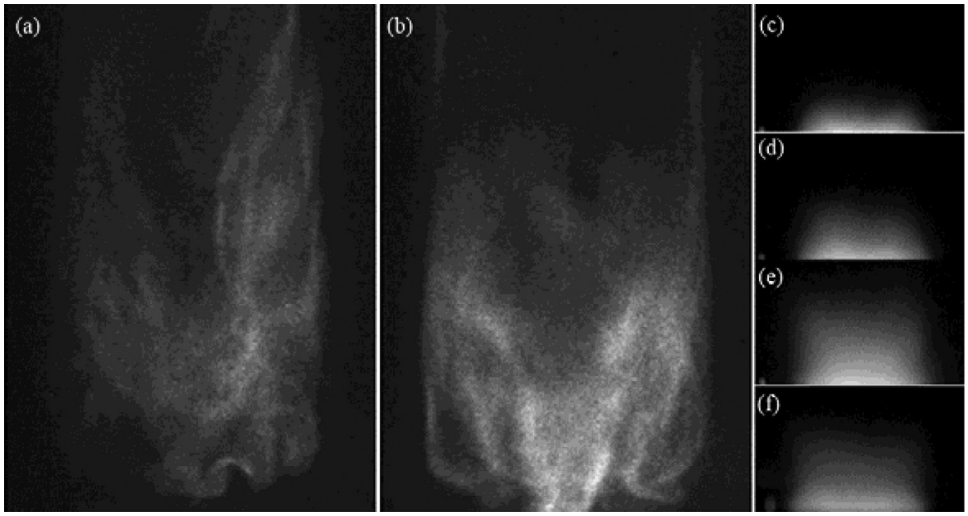

Fig. 3. $NH_2{}^*$ chemiluminescence: (a) without plasma, ϕ=0.94; (b) - (f) with plasma. Specifically, (b) ϕ=0.94 with V = 15 kV and f = 4 kHz (39 W); (c) ϕ=0.33 with V = 15 kV and f = 4 kHz; (d) ϕ=0.48 with V = 15 kV and f = 4 kHz; (e) ϕ=0.33 with V = 15 kV and f = 6 kHz (59 W); (f) ϕ=0.33 with V = 11 kV and f = 11 kHz (59 W).

creased to a regime where no flame could exist, it can be clearly seen from Fig. 3(c) to (f) that intensity of $NH_2{}^*$ chemiluminescence increases with the increase of both ammonia concentration, discharge power and discharge voltage. Without ammonia (ϕ=0), no $NH_2{}^*$ chemiluminescence (emission near 632 nm) is observed in the air plasma. The increase of $NH_2{}^*$ chemiluminescence (indicating an increase of kinetic effect) with both discharge power and voltage is consistent with the results shown in Fig. 2.

4. Conclusion

Plasma-assisted ammonia combustion is investigated experimentally for the first time. Plasma can simultaneously extend the lean blowoff limits of ammonia flames and reduce NO_x emission. The benefit of simultaneous flame enhancement and NO reduction may enable plasma-assisted ammonia/air combustion as a new direction for renewable, clean energy. The interaction between plasma and ammonia flames is new and largely unknown, requiring further study.

Declaration of Competing Interest

None.

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References

[1] H. Kobayashi, A. Hayakawa, K.D.K.A. Somarathne, E.C. Okafor, Science and technology of ammonia combustion, Proc. Combust. Inst. 37 (2019) 109–133.
[2] J. Choe, W. Sun, Blowoff hysteresis, flame morphology and the effect of plasma in a swirling flow, J. Phys. D Appl. Phys. 51 (2018) 365201.
[3] A. Shioyoke, J. Hayashi, R. Murai, N. Nakatsuka, F. Akamatsu, Numerical investigation on effects of nonequilibrium plasma on laminar burning velocity of ammonia flame, Energy Fuels 32 (2018) 3824–3832.
[4] G. Faingold, J.K. Lefkowitz, A numerical investigation of NH_3/O_2/He ignition limits in a non-thermal plasma, Proc. Comb. Inst. (2020) In Press, doi:10.1016/j.proci.2020.08.033.
[5] D. Pugh, J. Runyon, P. Bowen, A. Giles, A. Valera-Medina, R. Marsh, B. Goktepe, S. Hewlett, An investigation of ammonia primary flame combustor concepts for emissions reduction with OH^*, $NH_2{}^*$ and NH^* chemiluminescence at elevated conditions, Proc. Comb. Inst. (2020) In Press, doi:10.1016/j.proci.2020.06.310.
[6] V.M. Donnelly, A.P. Baronavski, J.R. McDonald, ArF laser photodissociation of NH_3 at 193 nm: internal energy distributions in NH_2 $\tilde{X}X^{\sim 2}B_1$ and $\tilde{A}^2A_1$, and two-photon generation of NH $A^3\Pi$ and $b^1\Sigma^+$, Chem. Phys. 43 (1979) 271–281.
[7] Y. Yi, X. Wang, A. Jafarzadeh, L. Wang, P. Liu, B. He, J. Yan, R. Zhang, H. Zhang, X. Liu, H. Guo, E.C. Neyts, A. Bogaerts, Plasma-catalytic ammonia reforming of methane over Cu-based catalysts for the production of HCN and H_2 at reduced temperature, ACS Catal. 11 (2021) 1765–1773.
[8] H. Zhong, X. Mao, A. Rousso, C. Patrick, C. Yan, W. Xu, Q. Chen, G. Wysocki, Y. Y. Ju, Kinetic study of plasma-assisted n-dodecane/O_2/N_2 pyrolysis and oxidation in a nanosecond-pulsed discharge, Proc. Comb. Inst. (2020) In Press, doi:10.1016/j.proci.2020.06.016.
[9] J.A. Miller, M.J. Pilling, J. Troe, Unravelling combustion mechanisms through a quantitative understanding of elementary reactions, Proc. Comb. Inst. 30 (2005) 43–88.
[10] R. d'Agostino, F. Cramarossa, S. De Benedictis, G. Ferraro, Kinetic and spectroscopic analysis of NH3 decomposition under R.F. plasma at moderate pressures, Plasma Chem. Plasma Process. 1 (1) (1981) 19–35.