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SUSTAINABLE ENERGY

Plasma-assisted Ammonia Combustion: The Effect of Equivalence Ratio on NO Emissions

As a carbon-free fuel and hydrogen carrier, ammonia has gained increasing attention in the combustion community to support the global decarbonisation efforts. However, it has unfavorable flame properties and emissions characteristics such as low flammability and intrinsic trade-off between nitric oxide emissions and slip ammonia that hinders its widespread integration to the heat and power sectors. As a promising technology, non-thermal plasma has already presented the potential to assist swirling ammonia flames with significant improvements in NOX abatement and flammability, while eliminating the need for blending with other reactive fuels such as hydrogen and methane. For this study, a plasma burner was designed to explore plasma discharge interaction with a highly swirling premixed ammonia flame over a range of equivalence ratios, and its effect on NO emissions. The results revealed that average plasma power varied with the equivalence ratio and the reduced electric field affected the NO emissions characteristics of premixed ammonia flames.

Keywords:
Ammonia, plasma-assisted combustion, NO emissions, swirling flames.

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INTRODUCTION

As a carbon-free fuel and hydrogen carrier, ammonia (NH_3) has the potential to support the clean energy transition in heat and power industries. Relatively easy to transport and store, NH_3 can also be regarded as a promising energy storage medium to leverage renewable energy sources in the electrical power generation mix. When produced from renewable energy, it provides a zero-carbon energy source to power gas turbines. However, burning NH_3 in gas turbines is a challenging task owing to the fundamental flame properties and emissions signature of NH_3, such as the low burning rate, high ignition energy and high tendency of elevated nitrogen oxides (NO_x) emissions[1,2]. The strategies for burning ammonia within the regulated NO_x emissions limits are therefore highly desirable and have become a key technological enabler to unleashing the vast potential use of ammonia for sustainable clean energy.

Co-firing ammonia with high-reactive fuels such as hydrogen is a proven strategy that can overcome the technical issues regarding the low burning rate while releasing zero-carbon emissions. As has been shown previously, ammonia flames doped with hydrogen (H_2) could conserve potential threats of flame stability and higher NO_x emissions depending on H_2 ratio in the blend and equivalence ratio (ϕ)[3]. Another potential strategy is the partial cracking of ammonia to hydrogen in a pre-combustion process. However, partially cracked NH_3 mixtures can produce an additional N_2 diluent component relative to NH_3-H_2 blends that reduce the efficacy of the burning rate enhancement and can elevate NO_x emissions with cracked H_2 fraction[4]. A new strategy is required that can simultaneously improve the fundamental flame properties and emissions performance of ammonia.

Plasma has emerged as a promising strategy with a proven track record of combustion enhancement, emissions abatement and fuel reforming, thanks to its unique chemistry that produces a rich pool of ions, electrons, other charged particles, neutral species, and heat[5] . Despite the relatively rich literature on plasma assisted hydrocarbon flames[5], the research on plasma-assisted ammonia combustion has lately started burgeoning with successful demonstration of applications for improved combustion and emissions mitigation[6–12]. The experimental studies [6–8,12] confirmed that plasma discharge has significantly extended the flammability limits of premixed swirling NH_3-air flames and reduced the NO emissions. However, the reduction has remained limited due to a series of factors such as plasma method, discharge gas, applied frequency and amplitude, and equivalence ratio.

In view of the above considerations, the aim of the research herein was to investigate plasma discharge-ammonia flame interactions over a range of equivalence ratios, with the primary focus on analysing discharge signal characteristics and its effects on NO emissions.

MATERIALS AND METHODS

The generic plasma burner used in the present work is based in Cardiff University's Gas Turbine Research Centre (GTRC) and shown schematically in Fig.1(a). The swirl manifold consists of eight tangential air supply ports of 3.5 mm ID, imparting a range of swirling motions to the upstream annular flow. The intensity of swirling motions can be quantified by a non-dimensional geometric swirl number given by [13]:

$$S_g = \frac{\pi r_0 D_0}{2A_t}\left\{\frac{m_\theta}{m_\theta + m_A}\right\}^2 \quad (1)$$

where r_o is the radius of the annular pipe into which the tangential flow is injected. D_o is the diameter of the annular pipe at the throat. The terms, m_θ and m_A, are the mass flow rates of tangential and axial inlets, respectively. A_t is the total cross-sectional area of all tangential inlets. In the setup, the flame is confined into a quartz tube of 100 mm ID.

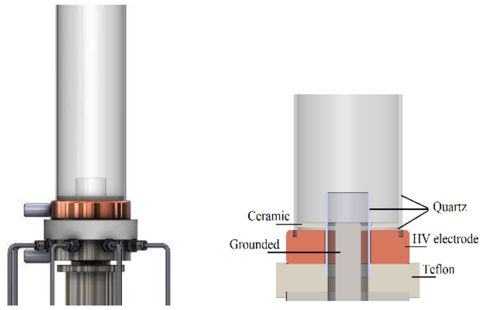

Fig.1. Plasma burner equipped with a coaxial DBD plasma generation system (a) CAD drawing and (b) cut-through view.

As shown in Fig.1 (b), a coaxial dielectric barrier discharge (DBD) plasma generation system was integrated to the swirl burner to produce volumetric non-thermal plasma in a discharge gap of 7.5 mm between two electrodes separated by a quartz nozzle of 40 mm ID serving as a dielectric or an insulator. A donut-shaped copper electrode received high voltage (HV) pulses while the stainless-steel swirl manifold was connected to the central stainless steel electrode of 25.4 mm which was connected to ground. The HV electrode was isolated from the grounded swirl manifold by a Teflon insulator. Before being applied to the discharge gap, a signal was first generated and then amplified using a waveform generator (Rigol 4000 series) and a high voltage amplifier (Trek 30/20), respectively. A HV probe (North Star PVM-5) and a current pulse transformer (Stangenes current pulse transformer) were used in combination with a high-speed oscilloscope (Lecroy HDO 6000) to monitor the discharge voltage and current waveforms included to measure the DBD pulses in the discharge gap. The electrical configuration of the rig is illustrated in Fig.2.

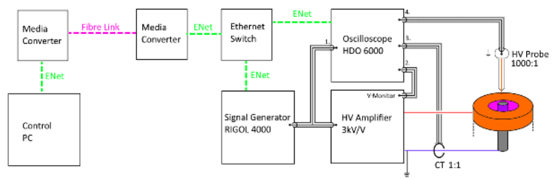

Fig.2. Electrical configuration of PAC tests at GTRC (burner detail omitted for clarity).

Gaseous emissions were sampled downstream of the confinement quartz tube using a 9-hole equal-area probe, regulated with a heat exchanger to maintain temperature at 433K. The gas extraction system includes a pump, filter block and sample lines maintained at the same temperature conforming to the specifications in ISO-11042[14].
The sample gas was split and delivered to multiple gas analysers housed in a single cabinet. NO concentrations were measured as hot and wet at 1 Hz using using heated vacuum chemiluminescence (Signal 4000VM), whereas dry O_2 concentrations were quantified by using a paramagnetic analyser (Signal 9000MGA).

Chemiluminescence spectrum of ammonia flames was scanned by using a UV/visible-capable spectrometer (Stellernet Inc BLUE-Wave) with a 100-mm focal length and a 25-µm wide entry slit which was coupled with a UV/visible-capable optical fiber head (Stellernet Inc DLENS with F600 fiber optic cable). The spectrometer incorporates 600-grooves/mm grating and a Si-CCD detector (Sony ILX511b) with a spectral resolution of 0.5 nm. The detector›s exposure time was set to 1 s and 20 scans were averaged to improve the signal-to-noise ratio (SNR).

In this study, the experiments were performed for premixed swirling NH_3-air flames at equivalence ratios ranging from 0.8 to 1.2. For each case, the applied voltage was kept constant at 21 kVpk and 400 Hz which would reliably maintain a plasma over the measurement duration. The geometrical swirl number was maintained constant at 4.36 ±0.012 across the equivalence ratio range by varying the ratio of m_θ/m_A. Thermal power output of the ammonia flames was kept constant at 8 kW. In all the test points, the inlet mixture stayed at ambient temperature; no preheating was employed to sustain flame in the combustion zone.

RESULTS

Figure 3 shows the phase resolved DBD energy derived from the acquisition of 10,000 current pulses in discrete time windows for the equivalence ratios of 0.8, 1.0 and 1.2, respectively. Phase-resolved discharge colour map indicates the density of pulses occurring at a given energy and phase on the AC cycle. The figure shows that the individual pulse energy did not exceed 0.2mJ, and DBD occurred on both the positive and negative-going rising edge of the voltage, giving the total energy of 1.825 J, 1.588 J and 1.416 J for equivalence ratios of 0.8, 1.0 and 1.2, respectively.

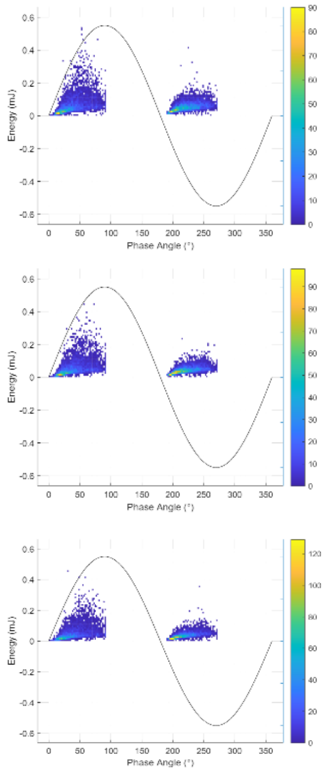

Fig.3. Phase-resolved pulse energy as a function of Equivalence Ratio, for P=8kW, V=21kVpk; (a) φ=0.8 (top), (b) ϕ=1.0 (middle), and (c) ϕ=1.2 (bottom).

Figure 4 shows the average plasma power, P_{avg} (over 10,000 pulses) as a function of the equivalence ratio. There was a notable reduction in P_{avg} with ϕ, and this behavior merits further investigation.

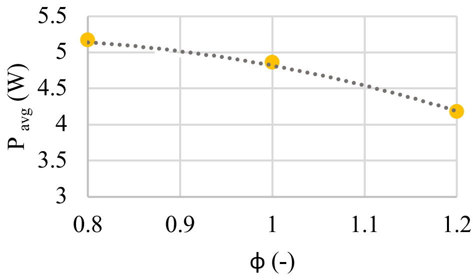

Fig.4. Average plasma power dependency on equivalence ratio.

Figure 5 shows the local variation of the reduced electric field, E/N in the discharge gap (E/N, the electric field (V/m) divided by the gas number density(1/m3) and given in the unit Townsend (Td) [5]) The calculated reduced electric field was varied in a range of 80-130 Td (1Td=10^{-21} V*m^2), regardless of equivalence ratio.

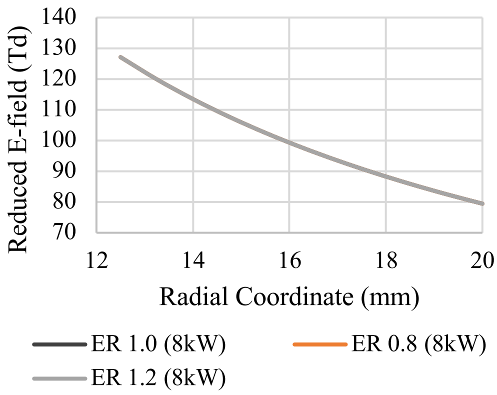

Fig.5. Reduced electric field in the space between the grounded central electrode and dielectric barrier. Note that E/N is nearly identical for all the equivalence ratios, which is expected given the fixed gas pressure.

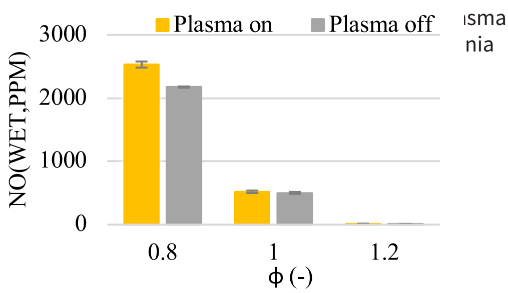

Fig.6. Comparison of NO emissions for plasma-assisted ammonia flames and conventional ammonia flames.

At fuel rich conditions, the plasma effect on NO emissions was negligible yet still more pronounced at the lean conditions, yielding higher NO concentrations than conventional ammonia flames. Regardless of the plasma activation, NO emissions notably abated at fuel rich conditions.

Figure 7 illustrates the peak intensity value of NH_2* excited radicals at 630 nm (corresponding to NH2 α band[15]) in the chemiluminescence spectrum of ammonia flames.

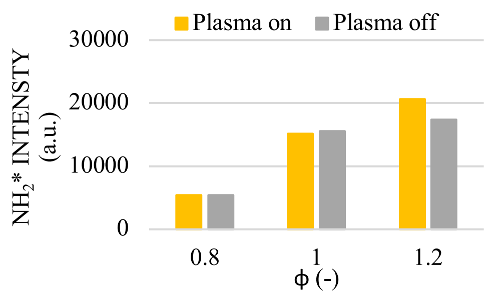

Fig.7. NH_2* chemiluminescence intensity of plasma-assisted ammonia flames and conventional ammonia flames over the equivalence ratio range.

In general, the radical concentration increased with the equivalence ratio indicating the decomposition of excessive ammonia in the flame region. The plasma discharges enriched the radicals in the ammonia rich flames. However, the effect was diminished at fuel-lean conditions.

DISCUSSION

In conventional ammonia flames, NO consumption at fuel rich conditions is enhanced through the reactions between NH_2 with NO to form either N_2 or NNH, both of which are the key reaction pathways of Thermal De-NO_x[1]. The increase in NH_2* chemiluminescence with equivalence ratio confirmed the reduction in NO concentrations as previously proposed in [6,16]. At lean conditions, NO formation is promoted through oxidization of NH and NH_2 to NO, mainly over the HNO intermediate [1]. However, the plasma discharge had no significant effect on NO emissions at rich conditions, even though yielding higher NH_2^* chemiluminescence. On the other hand, at lean conditions, the plasma effect became more stand-out, increasing NO emissions in the ammonia flames. The results are incongruent with others' findings [6,7] in which the plasma discharge ensured curbing NO emissions significantly in ammonia flames. This discrepancy might be related to the ratio of discharge power to thermal load. In the present study, this ratio was almost one third of the value in [6] , corresponding to only 0.063% of the thermal load at φ=1.0. Moreover, the E/N was varied in a range of 80-130 Td. As has been reported by other studies [10,17], for the E/N ≤100 Td, the main fraction of the plasma energy goes into the vibrational excitation of N_2, that does not produce effective plasma species to functionise ammonia flames. As the electric field varies with discharge gap, applied voltage and geometry of the electrode [17], the future study will focus on repeating the experiments at higher electric fields.

CONCLUSIONS

The effect of plasma discharge-ammonia flame interactions on NO emissions were experimentally investigated varying equivalence ratio at a constant thermal power output. NO emissions were compared against NH_2^* chemiluminescence intensity.

There is an apparent relationship between equivalence ratio and average plasma power, with lower equivalence ratio resulting in a measurably more energetic plasma.

In this study plasma discharges did not improve the NO emissions performance of premixed ammonia flames.

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Conflicts of interest
The authors declare no conflict of interest.

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