EMISSIONS PERFORMANCE OF STAGED PREMIXED AND DIFFUSION COMBUSTOR CONCEPTS FOR AN NH\textsubscript{3}/AIR FLAME WITH AND WITHOUT REACTANT HUMIDIFICATION

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ABSTRACT

Renewably generated ammonia offers a form of carbon-free chemical energy storage to meet the differences between uncertain supply and fluctuating demand and has the potential to support future energy requirements. The storage/transportation characteristics of NH\textsubscript{3} are favorable compared with H\textsubscript{2}, however there are combustion research challenges to enhance fuel reactivity whilst reducing harmful emissions production. The purpose of this work was to evaluate different fuel delivery concepts for a representative GT combustor. An experimental and numerical comparison was made between swirl-stabilized premixed and diffusion NH\textsubscript{3}-air flames at elevated inlet temperature (473 K). The exhaust NO\textsubscript{x} and NH\textsubscript{3} emissions generated from each concept were quantified to optimize combustor performance. High-speed OH\textsuperscript{*} and NH\textsubscript{2}\textsuperscript{*} chemiluminescence was employed to characterize the change in flame topology with variation in fuel-air equivalence ratio, and the resultant influence on measured emission concentrations. Chemiluminescence intensities were shown to elucidate changes in sampled exhaust emissions, enabling detailed analysis of intermediate chemistry. A comparison was made between experimental data and kinetic simulations, demonstrating the sensitivity of NO\textsubscript{x} emissions to premixed fuel-air equivalence ratio. A comparison was also made between exclusive primary airflow, and the staged introduction of secondary air, to quantify the change in NO\textsubscript{x} production between each configuration and improve fuel burnout. Secondary air loadings were incrementally increased through the combustor. Finally, reactant humidification was employed as a secondary process for NO\textsubscript{x} reduction, having shown favorable performance with NH\textsubscript{3}-H\textsubscript{2} mixtures, with the efficacy compared for both premixed and diffusion configurations.

Keywords: Ammonia, staged combustion, NO\textsubscript{x} emissions, humidification, chemical kinetics modelling.

NOMENCLATURE

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Definition</th>
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<tbody>
<tr>
<td>AFT</td>
<td>Adiabatic Flame Temperature (K)</td>
</tr>
<tr>
<td>CRN</td>
<td>Chemical Reactor Network</td>
</tr>
<tr>
<td>CRZ</td>
<td>Central Recirculation Zone</td>
</tr>
<tr>
<td>FZ</td>
<td>Flame Zone</td>
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<tr>
<td>ORZ</td>
<td>Outer Recirculation Zone</td>
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<tr>
<td>P</td>
<td>Pressure (MPa)</td>
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<tr>
<td>PFR</td>
<td>Plug Flow Reactor</td>
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<tr>
<td>Re</td>
<td>Reynolds Number</td>
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<tr>
<td>SA</td>
<td>Secondary Air</td>
</tr>
<tr>
<td>( \text{S}_g )</td>
<td>Geometric swirl number</td>
</tr>
<tr>
<td>( \text{S}_L )</td>
<td>Laminar Flame speed (cm·s\textsuperscript{-1})</td>
</tr>
<tr>
<td>T</td>
<td>Temperature (K)</td>
</tr>
<tr>
<td>WL</td>
<td>Water Loading (g·s\textsuperscript{-1})</td>
</tr>
<tr>
<td>( \Phi )</td>
<td>Equivalence Ratio</td>
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<tr>
<td>( \Phi_g )</td>
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1. INTRODUCTION

There is increasing interest in using renewably generated NH₃ (sometimes termed ‘green ammonia’) as a prospective energy carrier [1,2] and a mechanism for storing H₂. Green H₂ is produced via the electrolysis of water using renewable electricity, and in-turn green ammonia is manufactured using green H₂ and nitrogen. With existing infrastructure and favorable storage/transportation characteristics, the potential exists to supply power systems with NH₃, avoiding conversion back to H₂. However, this presents several combustion research challenges [2-6]: NH₃ reactivity is poor compared to traditional hydrocarbon fuels, with peak laminar flame speeds (Sₐ) at atmospheric conditions ~8 cm·s⁻¹. Combustion performance can be enhanced by thermolysis to form H₂ [7-9], preheating, or supplementary fuel blending [8-12]. A more significant challenge is the propensity for excessive exhaust NOₓ concentrations from fuel-bound nitrogen [12-15]. Conventionally employed lean, low-temperature combustor strategies are ineffective for NOₓ reduction with NH₃/air flames, with previous studies demonstrating concepts such as humidification [3] and staging [14-15] more successful, alongside an increase in combustor pressure [3,16-17]. This makes the prospective application of NH₃ for energy conversion more favorable for gas turbines.

1.1 Scope of This Work

Contemporary research is mostly directed towards the development of low-NOₓ NH₃ combustors [3,14,17], exploring staged concepts, and comparing performance against CH₄-air flames [15,18]. Through the optimized introduction of staged secondary-air, pathways have been identified that limit NOₓ production in a fuel-rich primary flame, whilst improving combustor efficiency through reducing unburned NH₃ and H₂ [3,14,17,18]. However, the number of detailed experimental studies with NH₃ flames remains limited, particularly at elevated inlet conditions, and an optimized flame configuration that provides both low NOₓ and NH₃ will greatly impact the performance of downstream staging. Recent work undertaken by the authors has focused on H₂ blending strategies [5], and changes in combustor pressure [3]. However, the purpose of this study was to expand on recent findings to investigate the change in emissions from a pure NH₃/air flame, exploring NO reduction pathways through the detailed numerical and experimental investigation of distinct mechanisms of fuel delivery. A fuel-flexible generic swirl burner was employed in both premixed and diffusion configurations with swirl-stabilized NH₃-air flames. Regions of excited OH* and NH₂* were visualized in the primary flame using Abel transformed high-speed chemiluminescence and compared against emissions produced. Changes in secondary air loading and reactant humidification were subsequently made to quantify any resultant variation in total emissions production.

2. EXPERIMENTAL CONDITIONS

Initial experiments were undertaken using NH₃-air mixtures with a fully premixed flame configuration, achieved by directing all reactants to the outer swirling flow (flow-path 2 - Fig.1). The fuel supply was then withdrawn for the non-premixed experiments, with NH₃ instead directed to the central diffusion jet (flow-path 1 - Fig.1). Data points were specified for primary equivalence ratios in the range Φ=0.9–1.2 by varying the swirling air flow rate. Ambient combustor inlet conditions of temperature (T) and pressure (P) were held constant at respective values of 473±5 K and 0.11 ±0.005 MPa. With the fully premixed configuration, swirler nozzle bulk outlet velocities were varied from ~9-11 m·s⁻¹ across the specified Φ range with Reynolds numbers increasing from Re=10,700 to ~13,400 at the highest flow conditions. These values were reduced by approximately 20% for the diffusion case. Initially, data were captured from only the primary flame (SA₁), with secondary airflow then increased at two fixed mass loadings: 3 g·s⁻¹ (SA₂) and 6 g·s⁻¹ (SA₃). These flow rates correspond to approximately 50 and 100% of primary airflow at the richest condition. Finally, humidification of the reactant airflow was appraised, having previously shown to be effective in reducing exhaust NOₓ concentrations with an NH₃/H₂ flame [3]. Limited steam flow rates could be achieved before the flame became unstable – realised by intermittent detachment from the burner nozzle – with data captured for a constant water loading of 0.2 g·s⁻¹ (WL₁) at Φ=1, corresponding to ~3%vol of the total reactant composition, or ~5%vol of the airflow. Fuel flow was maintained at a constant mass flow rate of 1.34 g·s⁻¹ for all conditions, to give a net thermal power of 25 kWth. The experimental conditions and flowrates are summarized in Table 1.

### TABLE 1: SUMMARY OF EXPERIMENTAL CONDITIONS

<table>
<thead>
<tr>
<th>Φ</th>
<th>Fuel Flow (g·s⁻¹)</th>
<th>Air Flow (g·s⁻¹)</th>
<th>SA (g·s⁻¹)</th>
<th>WL (g·s⁻¹)</th>
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<tr>
<td>0.9</td>
<td>1.34</td>
<td>9.04</td>
<td>0, 3, 6</td>
<td>0</td>
</tr>
<tr>
<td>1</td>
<td></td>
<td>8.13</td>
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<td>1.1</td>
<td></td>
<td>7.39</td>
<td>0</td>
<td></td>
</tr>
<tr>
<td>1.2</td>
<td></td>
<td>6.78</td>
<td>0</td>
<td></td>
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</table>

3. EXPERIMENTAL FACILITY

The employed swirl burner is presented as a cross-sectional schematic in Fig.1, with a central diffusion lance (Fig.1a) added to facilitate the introduction of a jet to the annular swirling flow. The lance ID (15 mm) was specified to generate approximately equal jet velocities (flow-path 1 in Fig.1) to the outer flow (flow-path 2) at the highest airflow condition. For the premixed configuration, reactants were mixed prior to entering the inlet plenum (Fig.1b) with flows metered using Coriolis mass-flow control (±0.35%). The plenum body was preconditioned to 473K using electric preheaters and compressed air, dried to a dew point of -17°C. System temperatures were allowed to stabilize for at
least an hour before experiments began. From the premix chamber (Fig.1c), reactants flow through a radial-tangential swirler (Fig.1d) with a geometric swirl number of $S_g = 0.8$ and out the exit nozzle ($r = 20\text{mm}$). Quartz windows (Fig.1e) provided optical access into the pressure casing (Fig.1g) used to enclose the secondary airflow which was introduced through a single inlet (flow-path 3 - Fig.1). For humidified experiments, water supply was regulated using another scaled mass-flow controller ($\pm 0.2\%$) and vaporized in heaters prior to entering the preheated air stream. Chemiluminescence measurements were taken perpendicular to the reactant flow direction through the optical windows, specified to give enhanced UV transmission. The primary flame was housed within a cylindrical quartz confinement tube (Fig.1f) at an expansion ratio of 2.5 from the nozzle. Combustor pressure was controlled using a water-cooled back-pressure valve with incremental control. Greater detail of the facility can be found in other publications [19-20].

**FIGURE 1**: CROSS-SECTIONAL SCHEMATIC OF BURNER, CASING ASSEMBLY AND SAMPLE PROBE (LABELS [not to scale])

### 3.1 Emissions

Exhaust gas emissions were sampled downstream of the quartz confinement and secondary air mixing zone using a 9-hole equal-area probe, water-conditioned with a heat exchanger to regulate sample temperature (433K) following specifications in ISO-11042 [21]. The pump, filter block and sample lines were also maintained at the same temperature. Total NOx (NO and NO2) concentrations were quantified using heated vacuum chemiluminescence as the measurement technique (Signal 4000VM). \( \text{NH}_3 \) measurements were made using the same analyzer and redirecting sample through a NO converter (Signal 410) to measure unreacted concentrations (81% conversion efficiency). All \( \text{NH}_3 \) and NOx concentrations were measured hot/wet and normalized to equivalent dry conditions using equation (9) in ISO-11042. Dry \( \text{O}_2 \) measurements were made using a paramagnetic analyzer (Signal 9000MGA) and used to subsequently normalize NOx/\( \text{NH}_3 \) readings to 15% \( \text{O}_2 \) equivalent (equation (10) in ISO-11042). At each experimental condition, burner temperatures, pressures, flows and emissions were monitored and, once stable, held for a minimum of 60 samples to be taken. Systematic uncertainties in the analyzer (linearisation and span gas certification), were combined with any fluctuations in measurement to give the total uncertainty represented by the plotted error bars.

### 3.2 Chemiluminescence

Chemiluminescence measurements were obtained at each experimental condition, targeting electronically excited intermediate chemical radicals within the \( \text{NH}_3 \)-air flames. The species investigated were \( \text{OH}^* \) and \( \text{NH}_2^* \), captured using a combination of Phantom v1212 high-speed CMOS camera, Specialised Imaging SIL40HG50 high-speed image intensifier, UV lens (78 mm, f/11), and narrow bandpass filters selected specifically for each species (further information on this facility is found in other works [22]). \( \text{OH}^* \) chemiluminescence images were taken focusing specifically on the well-known \( \text{A}^2\Sigma^+ - \text{X}^2\Pi \) \( \text{OH}^* \) system [23] using a 315 nm ($\pm 15$ nm FWHM) bandpass filter. Few experimental studies have conducted \( \text{NH}_2^* \) chemiluminescence measurements, and these have typically been confined to canonical reactors [24-27] or shock tubes [28-29] rather than \( \text{NH}_3 \) flames, where \( \text{NH}_2 \) and NH intermediate chemistry is key to \( \text{NH}_3 \) oxidation [1-3]. For \( \text{NH}_2^* \) measurements, a single peak of the \( \text{NH}_3 \) a band was selected at 630 nm [23,26] using a bandpass filter centered at 632 nm (±10 nm FWHM). Images were captured at 4000 Hz, with the image intensifier gated at 10$\mu$s and the intensifier gain held constant throughout. The image resolution is 4.6 pixels/mm, resulting in a field of view of 75 mm (axial, y) by 100 mm (radial, x) relative to the edge and centerline of the burner exit nozzle, respectively (Fig. 2). Each instantaneous chemiluminescence image was filtered using a 3x3 median filter and temporally averaged from 2000 images ($t = 0.5$ s). The resultant image was then background corrected and transformed using a modified Abel inversion algorithm. This provided an axisymmetric planar representation of the electronically-excited radical species distribution within the swirling flow field [30]. A comparison is shown between the averaged raw \( \text{NH}_2^* \) chemiluminescence (Fig. 2a) and equivalent Abel transform (Fig. 2b) for an example case in Fig. 2 where the centerline of the burner nozzle is represented by $x = 50\text{mm}$ and flow enters from the bottom.
4. CHEMICAL REACTOR NETWORK MODELING

The chemical kinetics of the premixed experimental burner were simulated using Chemkin-Pro. PREMIX was employed with the laminar flame speed calculator to model changes in $S_L$ with $\Phi$. Solutions were based on an adaptive grid of 1000 points, with multi-component transport properties. The equilibrium reactor was used to generate adiabatic flame temperatures (AFT) under conditions of constant enthalpy/pressure. The NH$_3$ flame model employed the Okafor et al. reaction mechanism, optimized for use with CH$_4$/NH$_3$-air mixtures [18] with 59 chemical species and 356 reactions. The predicted changes in $S_L$ (solid) and AFT (dashed) are shown for the range of experimental $\Phi$ in Fig. 3. A comparison is shown between the dry case at WL$_0$, and the humidified condition WL$_1$, alongside a CH$_4$-air flame at equivalent conditions (GRI mechanism [30], 53 species and 325 reactions). The limited water loading employed in this study was shown to provide a ~12\% reduction in NH$_3$-air $S_L$ across the specified $\Phi$ range, with both results markedly lower than the equivalent CH$_4$-air flame. As speeds were already low for the NH$_3$-air flame, water addition provided a significant drop in reactivity, leading to blow off instabilities when humidified fractions were further increased. AFT was shown to be high enough to facilitate thermal NO$_x$ formation for the NH$_3$-air flame and was reduced by ~50K at WL$_1$.

A chemical reactor network (CRN) model was also developed to simulate the chemistry of the experimental swirling NH$_3$-air flame [10-11, 24], analogous to the simulation previously presented by the authors [3]. Inlets were used to provide fuel/air/steam flows, with three perfectly stirred reactors (PSR) to model the pre-mixing, flame and central recirculation zones (CRZ) (20\% recycled flow [25]). The outlet from the flame zone (FZ) fed a Plug Flow Reactor (PFR) to simulate reactions in the post-flame zone. The model structure is outlined in Fig. 4 and was established with representative combustor geometry and residence times calculated from empirical flow conditions. Initial reactor temperatures were defined from equilibrium calculations. Heat loss was estimated to be 10\%, with initial values compared to empirical results at dry conditions to give favorable agreement with sampled concentrations. It should be emphasized that moderate uncertainty is associated with CRN modelling, and the system developed was primarily used for qualitative analysis of trends, rather than detailed prediction.

5. RESULTS

5.1 Premixed Flame

Measured NO (empty) and NH$_3$ (shaded) emissions from the fully premixed flame configuration are shown plotted against $\Phi$ in Fig. 5 – data are provided for SA$_0$/WL$_0$ only. The markers show the averaged experimental data normalized to dry 15\% O$_2$ conditions, with results from the reactor network model represented by the lines. Measured NO$_2$ fractions were negligible, with substantial concentrations only measured at $\Phi$=0.9 (<40ppmv). Overall, favorable agreement is shown in trends between the sampled and modelled results. The model
also allows for the simulation of exhaust H2 concentrations, which were not measured and are shown to rise sharply above Φ=1. This demonstrates that most of the fuel-bound nitrogen is liberated in the primary flame zone, and unreacted fuel is largely formed as H2. Therefore, fuel-NOx production will be limited in the secondary flame zones, though NH3 concentrations appear to rise around Φ=1.1.

A pronounced decrease in NO with increasing Φ is also evident in Fig. 5. This results from reduced thermal formation with the Zel’dovich mechanism, with diminished availability of OH for NH3 oxidation [16,18] and combination with HNO [3,5], alongside enhanced NO consumption with NH2 through the chain carrying reaction NH2+NO→NNH+OH, and the terminating reaction NH3+NO→H2O+N2 [3]. Figure 6 shows the change in OH* and NH3* Abel transformed chemiluminescence profiles with change in Φ. Half of the axisymmetric flame is shown, with x=0 mm corresponding to the burner nozzle centerline and flow from the bottom. The color maps in these images have been normalized to the maximum for each species dataset to emphasize the change in reactive intensity. A complementary trend is evident for change in Φ between the reduction of OH* intensity, an increase in NH3*, and a drop in measured exhaust NO concentrations. Using OH* as a generalized marker for heat release [23], the flame structure appears typical for a swirling premixed configuration [22,31], with the flame stabilized around a conical shear layer of zero axial velocity, with central and outer recirculation zones (CRZ and ORZ respectively). There appears to be increased reacting flow in the ORZ – giving a characteristic ‘M’ flame, compared to the ‘V’ observed for near equivalent CH4/air flames with this burner [31].

5.2 Diffusion Flame

A comparison between measured emission concentrations from the diffusion and premixed configurations is presented in Fig. 7 – again data are shown for SA0/WL0 with change in Φ. Lines are this time included for clarity as rather than presenting modelled data. There is a marked difference between the two cases, particularly at the leanest Φ, with the diffusion flame generating significantly lower NO concentrations – once again

![Figure 5](image1.png)

**Figure 5:** Measured and modelled NO (empty) and NH3 (shaded) emissions for the premixed NH3-air flame, alongside simulated H2 concentrations.

![Figure 6](image2.png)

**Figure 6:** Abel transformed OH* and NH3* chemiluminescence for the premixed NH3-air flame and increasing Φ (colormap normalized to species dataset maximum).

![Figure 7](image3.png)

**Figure 7:** Measured NO (dashed line) and NH3 (solid line) emissions for premixed and diffusion NH3-air flames.
NO\textsubscript{2} was measured to be negligible aside from \(\Phi=0.9\), with values similar to the premixed case. Local NH\textsubscript{3}-rich zones lead to the increased production of reactive NH\textsubscript{3} within the diffusion flame as observed in the NH\textsubscript{3}\textsuperscript{*} chemiluminescence shown in Fig. 8. Once again this suggests reactive NH\textsubscript{3} acts to consume NO through the kinetics discussed for the premixed case, with the more dispersed region of NH\textsubscript{3}\textsuperscript{*} leading to a drop in measured NO emission concentrations.

The colormap in Fig. 8 are this time normalized to the maximum intensity in each image to emphasize the change in flame topology with \(\Phi\). A comparison is shown between the raw averaged chemiluminescence (Fig 8b), and equivalent Abel transform (Fig 8c) - As bulk flow is decreased with increasing \(\Phi\), the effective swirl reduces and the flame appears to contract towards the centerline diminishing the influence of the CRZ, and extending beyond the observable window. Instantaneous images of measured NH\textsubscript{3}\textsuperscript{*} intensities are also included in Fig. 8a (captured at intervals of \(~0.05\text{s}\)) to highlight the transient structure of the diffusion flame (\(\Phi = 1\)) employed and more dispersed regions of reacting flow, facilitated through the use of the high-speed chemiluminescence.

![Image](image_url)

**FIGURE 8:** INSTANTANEOUS (a), AVERAGED RAW (b), AND ABEL TRANSFORMED (c) \(\text{NH}_3^*\) CHEMILUMINESCENCE FOR THE DIFFUSION \(\text{NH}_3\)-AIR FLAME (COLORMAP NORMALIZED TO IMAGE MAXIMUM).

Whilst the diffusion flame provides the advantage of lower exhaust NO\textsubscript{x} concentrations, combustor efficiency appears comparatively poor, with significantly higher unreacted NH\textsubscript{3} fractions observed in Fig. 7. Measured concentrations are shown to rise with increasing \(\Phi\) to provide an order of magnitude difference between the two flame configurations. This primarily results from the elevated NH\textsubscript{3} concentrations observed in Fig. 8, with the respective chain branching and third body reactions

\[
2\text{NH}_3 \leftrightarrow \text{NH}_3 + \text{NH} \quad \text{and} \quad \text{NH}_3 + \text{H} + \text{M} \rightarrow \text{NH}_4 + \text{M} \quad [3].
\]

Fuel-bound nitrogen in the excessive NH\textsubscript{3} emissions from the primary flame would therefore be expected to increase aggregate NO\textsubscript{x} production with the introduction of staged secondary air to improve combustor efficiency.

### 5.3 Secondary Air Introduction

Secondary air was introduced to the combustor at two constant mass loadings: 3 g\textsuperscript{\text{-1}} - SA\textsubscript{1}, and 6 g\textsuperscript{\text{-1}} - SA\textsubscript{2}. The change in global fuel-air equivalence ratio (\(\Phi_G\)) was calculated by adding the secondary air component to the primary loading, providing a range of globally lean experimental points. A comparison between the emissions produced from the diffusion and premixed flame configurations at SA\textsubscript{1} is shown in Fig. 9 (once again lines are included for clarity and do not represent modelled data). Emissions from the \(\Phi_G=0.67\) (corresponding to a primary \(\Phi=0.9\)) case appear to provide similar measured concentrations to the equivalent SA\textsubscript{0} experiment – as may be expected from a lean primary flame with minimal unburned fuel concentrations. However, once the primary \(\Phi\) is increased above stoichiometric, there is an offset between the measured concentrations for SA\textsubscript{1} and those presented in Fig 7. Unreacted H\textsubscript{2} and NH\textsubscript{3} are consumed in the secondary flame zone to produce more NO\textsubscript{x}. The order of magnitude difference in NH\textsubscript{3} concentrations from the primary diffusion case lead to a crossover at \(\Phi_G=0.83\), with higher NO values than the equivalent premixed condition. Here the optimal configuration is identified with both NO and NH\textsubscript{3} \(<75\text{ppmvd}\). The method of secondary air

![Image](image_url)

**FIGURE 9:** MEASURED NO (DASHED LINE) AND NH\textsubscript{3} (SOLID LINE) EMISSIONS FOR PREMIXED AND DIFFUSION NH\textsubscript{3}-AIR FLAMES WITH SECONDARY AIR LOADING (SA\textsubscript{1}).
introduction employed in this study is simple with comparatively poor mixing. This leads to inefficiency for the diffusion case, with significant unreacted NH$_3$ concentrations still measured at the richest condition and combustion reactions quenched in the emissions sample probe.

With the NH$_3$-air flame so sensitive to change in $\Phi$, it was essential to ensure the quartz confinement tube remained effectively sealed at the premix chamber (Fig 1c), with no secondary air entrainment into the primary flame-zone. Measured chemiluminescence intensities were used to validate the employed experimental setup. Figure 10 provides a comparison of the NH$_3$* chemiluminescence for an example premixed case – colormaps have been normalized to the maximum intensity measured across the 4-image dataset. A marked difference is observed between the $\Phi$=0.9 and 1 SA$_0$ images, with the maximum intensities around 29% higher in the latter. Whereas the respective differences observed for SA$_1$ and SA$_2$ cases were 2.4 and 0.7%, and hence the images appear near-identical in Fig. 10.

![Figure 10: Abel transformed NH$_3$* chemiluminescence for premixed NH$_3$-air flames with secondary air loading (colormap normalized to dataset maximum).](image)

Secondary air loading was increased further to SA$_2$, with the emissions dataset for the ostensibly optimal $\Phi$=1.2 plotted against air loading in Fig. 11. For the diffusion case, as combustor efficiency is increased with enhanced air loading, more NH$_3$ is consumed in the secondary zone leading to higher NO concentrations, with some unburned fuel remaining in the product sample. A marginal increase in NO was observed for the premixed flame from SA$_1$ to SA$_2$ possibly resulting from enhanced H$_2$ consumption, or greater O$_2$ concentrations facilitating more NO production through OH and HNO pathways, such as HNO+OH→NO+H$_2$O. NO$_x$ fractions were also shown to rise with an increase in secondary air loading; for low secondary air cases, negligible concentrations were measured for either flame configuration at $\Phi$=1.2. However, once secondary air loading was increased further to SA$_2$, premixed and diffusion primary flames gave respective measured NO$_2$ concentrations of ~10 and ~100 ppmvd. Again, this results from greater O$_2$ availability in the secondary flame-zone, with models suggesting enhanced third body production with NO through NO+O+M↔NO$_2$+M. The results therefore suggest that whilst combustor efficiency can be increased through the introduction of secondary air, the dilution must be carefully controlled, as the excessive introduction of air may lead to greater NO$_x$ production. Trials undertaken with CH$_4$-air mixtures in a similar burner configuration demonstrated an opposing trend [32]. Limited NO$_x$ sensitivity to primary flame equivalence ratio was observed as a function of thermal production, with the NH$_3$-air flame studied here shown to be more responsive. Increased secondary air loading was also shown to reduce NO$_x$ production, compared to the increase observed here.

![Figure 11: Measured NO (dashed line), NO$_2$ (dotted line) and NH$_3$ (solid line) emissions for premixed and diffusion NH$_3$-air flames with increased secondary air loading at $\Phi$=1.2](image)

5.4 Reactant Humidification

Humidification has previously been shown by the authors to be an effective mechanism for NO$_x$ reduction in NH$_3$/H$_2$ flames, dropping concentrations by an order of magnitude for reactant loadings of ~10% [3]. However, as discussed in previous sections, only limited steam loading could be achieved with the NH$_3$-air flame at 473K, before reactivity was diminished to the point of instability with intermittent extinction, or detachment from the burner nozzle. Whilst $S_L$ is shown to drop by 12% in Fig. 3, peak heat release rate is shown to reduce by ~25% from WL$_0$ to WL$_1$. It is therefore unsurprising that increasing water loading beyond this limited value leads to extinction. Nevertheless, the change in emissions production was quantified for the specified loading at $\Phi$=1 with the results presented in Fig. 12 for both the premixed and diffusion cases. There is an observed decrease in NO emissions with water loading for the premixed case, however this is small for the limited amount of H$_2$O introduced when compared with the efficacy for NH$_3$/H$_2$ mixtures. From CRN modelling, this reduction is simulated to
result from enhanced NH₃ consumption pathways 1) NH₂+NO ↔ NNH+OH, and 2) NH₂+NO ↔ H₂O+N₂, alongside a drop in thermal NO production. Figure 13 presents a comparison of the NH₂* chemiluminescence for the premixed case at different water loadings, with colormaps again normalized to image maximum to emphasize the change in topology. The influence of H₂O is subtle, but a marginal extension of the flame brush is observed, as would be expected from reduced reactivity, and indicates a larger region of NH₂*. For the diffusion case, minimal impact was observed with NO values marginally increasing, however this change is within the measured uncertainty of the experimental data. A rise in unreacted NH₃ was observed for both flame configurations, modelled to result from increased NH₂ production and could enhance NOₓ production with downstream staging. The results therefore suggest the suitability of humidification for NOₓ reduction is diminished for NH₃-air flames compared to NH₃/H₂ mixtures.

6. CONCLUSIONS

An experimental and numerical comparison was made between swirl stabilized premixed and diffusion NH₃-air flames at elevated inlet temperature (473 K). High-speed OH* and NH₂* chemiluminescence was employed to characterize the change in flame topology with variation in fuel-air equivalence ratio, and the resultant influence on measured emission concentrations. Chemiluminescence intensities were shown to elucidate changes in sampled exhaust emissions, enabling detailed analysis of intermediate chemistry.

Markedly different results were obtained for each primary flame configuration, with the diffusion case offering substantially lower measured NO concentrations at leaner conditions. The premixed flame gave more favorable emissions once an equivalence ratio of 1.2 was reached. The premixed flame also provided greater combustion efficiency, with NH₃ concentrations approximately an order of magnitude lower than each equivalent diffusion case, and unreacted fuel primarily formed as H₂. Favorable agreement was observed between experimental results, and data obtained from empirical chemical reactor network modelling. This facilitated the detailed chemical analysis of emissions formation.

The burner was subsequently employed in a staged configuration, with the incremental introduction of secondary air to increase combustor efficiency and reduce unburned fuel. Again, at the leanest global equivalence ratios, the diffusion flame provided superior emissions performance. However, for the richest experimental condition, the premixed primary flame provided NO and NH₃ concentrations <75 ppmvd. This resulted from a combination of low NO production in the primary flame and reduced fuel-bound nitrogen in the secondary reaction zone. The amount of secondary air loading must be carefully controlled however, as excessive oxygen can lead to the enhanced production of both NO and NO₂.

The efficacy of reactant humidification for NOₓ reduction was also appraised for the NH₃-air flame, having shown favorable performance with NH₃/H₂ mixtures. Limited H₂O concentrations could be achieved (3%vol of reactant composition) with the stable flame at the specified inlet temperature, before the drop in mixture reactivity led to blow off and extinction instabilities. A reduction in NO with H₂O increase was observed for the premixed case, however both flame configurations provided an increase in exhaust NH₃, which could lead to greater
NO production in a staged combustor and diminishing the suitability of humidification for NOx reduction.

A database of analytical results was generated, useful to the community for CFD validation, as increased work is undertaken with NH3 flames.

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