Influence of steam addition and elevated ambient conditions on NO\textsubscript{x} reduction in a staged premixed swirling NH\textsubscript{3}/H\textsubscript{2} flame

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Abstract

There is growing interest in the application of renewable NH$_3$ to support future energy requirements, however, combustion designs and strategies require considerable development to reduce NO emissions in particular. A turbulent premixed burner was used experimentally and numerically to appraise potential pathways for operational NO reduction with a premixed NH$_3$/H$_2$/air flame. Reactants were supplied at elevated temperature with parametric changes made to pressure and humidity. Favorable agreement was demonstrated between exhaust gas measurements and chemical kinetic simulations with a reactor network model, showing NO emissions to be sensitive to operational equivalence ratios, increasing by several orders of magnitude across the experimental range. The lowest NO concentrations were achieved at the richest conditions, accompanied by high unburned fuel fractions in the product stream. An increase in combustor pressure remarkably reduced exhaust NO concentrations primarily due to enhanced NH$_2$ formation, and subsequent NO consumption in the post-flame zone. Reactant humidification was explored in detail for the first time with this fuel, and shown to reduce NO production limiting thermal pathways with the extended Zeldovich mechanism. NO consumption in the post-flame zone was also enhanced through an increase in OH-produced NH$_2$, and together with pressure, resulted in elevated exhaust NH$_3$ concentrations. Whilst this effect was comparatively small, it meant that leaner humidified operation could be employed to reduce unburned fuel fractions without an NO penalty. Emissions performance was further improved by the application of staged combustion, with secondary air used to improve fuel burnout. Humidity and pressure were optimized in the staged configuration to achieve operation with sampled respective NO and NH$_3$ exhaust fractions of 32 and 50 ppmv (15% O$_2$), at a globally lean equivalence ratio.

There is considerable scope for further system optimization through improved mixing of secondary air and increased ambient pressure.

Keywords

Ammonia, Turbulent premixed flames, Humidified combustion, Staged combustion, Chemical kinetics.
1. Introduction

There is increasing interest in the application of 

NH$_3$ to support future energy requirements primarily due to its favourable transport and storage characteristics [1-2], with existing infrastructure particularly attractive for rapid deployment [3]. The potential exists to employ NH$_3$ directly as a fuel in modified power plants and avoid complete conversion back to hydrogen. However, there are several outstanding combustion challenges associated with the utilization of NH$_3$, principally due to its low flame speed, temperature and reactivity, in addition to the huge potential for NOx production [4-6].

Supplementary fuels such as H$_2$ have been shown to improve and stabilize operational performance [6-12]. Nevertheless, traditional combustor designs require fundamental appraisal and development to reduce NOx emissions before NH$_3$ can realistically be considered for sustained use in relatively clean and efficient technologies, such as gas turbines [8-11].

1.1 Scope

The aim of this work was to experimentally and numerically appraise the application of an NH$_3$/H$_2$ blend in a premixed swirl combustor at elevated temperature and pressure, to investigate potential pathways for operational NOx reduction. First, the change in formation is characterized for variation in equivalence ratio, with chemical kinetic processes simulated using a reactor network model. Change in operational NOx concentrations are then studied in detail at elevated conditions, increasing combustor pressure with density-scaled flow. This is combined with the humidification of premixed reactants to investigate the effect of H$_2$O on NOx formation for the first time with this fuel, whilst maintaining flame stability. Finally, the combustor is employed in a staged configuration to allow for the introduction of secondary air, similar to existing rich-burn, quick-quench, lean-burn (‘RQL’) systems [13]—which in typical applications operate at significantly higher pressures—while exploring parameter changes in flow, humidity and pressure with secondary air flow. Chemical kinetics influences are explored in detail with the empirical NH$_3$/H$_2$ flame and changes in pressure and humidity. The concepts investigated are combined to develop a combustor configuration that provides optimized emission performance.
Experiments were undertaken using a premixed swirling burner at Cardiff University's Gas Turbine Research Centre (GTRC) housed within an optical pressure casing, and modified from previous programs [14-15]. The burner and casing assembly are shown in Fig. 1. The instrumentation lance (Fig. 1a) facilitates condition measurements at the swirler outlet. Preheated reactants enter the inlet plenum (Fig. 1b), with mass-flow rates quantified using Coriolis meters (±0.35%). Gasous NH\textsubscript{3} and H\textsubscript{2} were supplied independently from external storage, with air provided from a compressor (GA-45 VS) dried to a dew point of -17°C (DPR 860). Steam was supplied using a heated line, with the burner conditioned to operational temperature using preheated air. Water supply was regulated using another mass-flow controller (±0.2%) and vaporized in two heaters. After entering the plenum, reactants travel through the premix chamber (Fig. 1c) to a single radial-tangential swirler (Fig. 1e) and out through the burner exit nozzle (20 mm radius). The outlet has a geometric swirl number equivalent to \( S_g = 0.8\). Optical access is afforded via quartz viewing windows (Fig. 1f) housed within the casing (Fig. 1g) with flame chemiluminescence imaged from above. The burner was operated with a cylindrical quartz confinement tube, at an expansion ratio of 2.5 from the burner nozzle exit (Fig. 1h). Secondary air for staged combustion is introduced through the casing inlet (Fig. 1d), flowing around the quartz tube. Additional transducers were installed to monitor operational parameters, with further detail provided elsewhere [14-15]. The system was pressurized using a water-cooled back-pressure valve with incremental control.

![Facility Schematic](image)

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**Fig. 1** Facility Schematic
2.1 Chemiluminescence

The chemiluminescence of an excited hydroxyl (OH) radical has been used to provide a non-intrusive indication of localized heat release, and a general marker of flame front location [14-16]. The image plane is centered on the burner exit, with respective view fields of ~100/75 mm in the radial and axial directions. For each experiment 200 images were captured at a rate of 10 Hz, with intensifier gain held constant. Intensities measured for each dataset were background-corrected and averaged, with a false colour map applied. As demonstrated in previous studies [14], the flow-field generated by the swirl configuration has been evaluated using particle image velocimetry [17], with the flame structure typical for a premixed swirl of this design: The outward flow generates a conical flame around a shear layer of zero axial velocity, and central recirculation zone (CRZ). A modified open-access Abel inversion algorithm [14] was used to transform the temporally-averaged images into planar representations of OH* chemiluminescence intensity distribution. Detailed information regarding the image capture system and processing technique can be found elsewhere [14-15].

2.2 Emissions Measurements

The emissions measurement system was designed in accordance with ISO-11042 [18]. Exhaust products were sampled downstream of the quartz confinement using a multi-point equal area probe connected to a water-conditioned heat exchanger, used to regulate the sample temperature to 433 K. A heated pump was used to deliver products to the analysis suite, and was also maintained at 433 K together with the filter and sample lines. NOx (NO + NO2) concentrations were quantified using a heated vacuum chemiluminescence analyzer (Signal 4000 V M). The sample stream could be redirected through an NH3 to NO converter (Signal 410) to measure unburned concentrations with an 80% conversion efficiency. All NOx and NH3 concentrations were measured hot/wet with captured data normalized to equivalent dry conditions (Eqn. (9) ISO-11042 [18]).
Additional sample was dried in a chiller before 
concentration was quantified using a paramagnetic 
analyzer (Signal 9000 MGA). Dry measurements were 
subsequently normalized to equivalent 15% O
subscript 2 (Eqn. (10) ISO-11042 [18]). Burner outputs were 
monitored after changing experimental set-points, and 
with temperatures stable, flows were held on condition 
and at least 60 samples taken. NO and NH
subscript 3 were measured separately, with richest 
test points requiring small amounts of air dilution in 
the sample to provide O
subscript 2 for NH
subscript 3 conversion and measurement. NH
subscript 3 concentrations were corrected to account 
for this dilution prior to analysis. Measured uncertainties 
comprise analyzer specification, linearization and span 
gas certification. These were combined with fluctuations 
in measurement to give a total uncertainty represented 
by the plotted error bars.
An NH\textsubscript{3} blend with an NH\textsubscript{3}/H\textsubscript{2} ratio of 70/30% mol was specified for this program. Alternative ratios were appraised \cite{11}, with the chosen blend shown to provide most stable performance between a susceptibility to flashback – apparent with excessive H\textsubscript{2} (> 40%) – and blowoff stability at richer conditions. This ratio has also been shown to behave analogously to a premixed CH\textsubscript{4} flame \cite{8}, with the employed burner characterized extensively using CH\textsubscript{4}/air mixtures \cite{19}. A comprehensive test matrix was specified and only the most significant results are presented and discussed, with the full dataset available as supplementary material.

Baseline conditions were specified at a thermal power of 25 kW with a combustor pressure of 0.105 MPa (P\textsubscript{1}), no supplementary steam addition or secondary airflow. Equivalence ratios were calculated relative to the stoichiometric fuel-air ratio of the NH\textsubscript{3}/H\textsubscript{2} blend (Ø= fuel-air/fuel-air\ stoich from 0.3 H\textsubscript{2} + 0.7 NH\textsubscript{3} + 0.675 O\textsubscript{2} + 2.54 N\textsubscript{2} \rightarrow 1.35 H\textsubscript{2}O + 2.89 N\textsubscript{2}) and were initially trailed in the range Ø= 1 - 1.4, with values Ø= 1.2, 1.25, and 1.3 (air flows of 6.56, 6.30, and 6.06 g∙s\textsuperscript{-1}) selected for detailed parametric study with independent changes in humidity, pressure, and secondary airflow.

Steam was introduced at four distinct humidified fractions: W\textsubscript{L1} = 0, W\textsubscript{L2} = 0.2, W\textsubscript{L3} = 0.4 and W\textsubscript{L4} = 0.6 g∙s\textsuperscript{-1} corresponding to overall reactant fractions of 0, 3.43, 6.63, and 9.63% mol at Ø= 1.2, which rise marginally due to the drop in airflow. Secondary air loadings for staged combustion were specified at ~50% and ~100% of the baseline combustor airflow (SA\textsubscript{1} = 0, SA\textsubscript{2} = 3 and SA\textsubscript{3} = 6 g∙s\textsuperscript{-1}). Pressure was increased across the range P\textsubscript{1} = 0.105, P\textsubscript{2} = 0.131, P\textsubscript{3} = 0.158 and P\textsubscript{4} = 0.184 MPa, with all given flow rates scaled with change in density to maintain equivalent nozzle exit velocities and residence times in the combustor. The experimental specification is summarized in Table 1, with a detailed matrix outlining each condition provided in the supplementary material, alongside measured NO\textsubscript{x} and NH\textsubscript{3} concentrations. The highest pressure specification was governed by the maximum sustainable NH\textsubscript{3} vapor flow of 1.8 g∙s\textsuperscript{-1}, with current operation limited at this point. At higher flows, NH\textsubscript{3} expansion reduced the drum temperature to limit supply vapor pressure. Combustor inlet temperature was maintained at 423 ± 5K.
Chemical Kinetics Modelling

Chemical kinetics were modelled for each experimental condition using CHEMKIN-P. The PREMIX reactor was initially used to provide 1-D simulations of changes in laminar flame speed (\(S_L\)).

Solutions were based on an adaptive grid of 1000 points, with multicomponent transport properties and trace species approximation. The equilibrium tool was used to generate adiabatic flame temperatures (AFT), under conditions of constant pressure and enthalpy.

Each simulation employed the Tian et al. reaction mechanism [20] comprising 84 chemical species and 703 reactions, and optimised for use with NH\(_3\) mixtures. Several other mechanisms were appraised [21-22], with the employed chemistry set providing the most favourable agreement with experimental data, along with the reduced mechanism developed by Xiao et al. [22]. The predicted changes in \(S_L\) and AFT are shown for a range of \(\phi\), with the highest pressures and H\(_2\) loadings in Fig. 2 (changes in AFT with pressure are not shown as differences were negligible). At baseline conditions AFT is high enough to facilitate significant thermal NO\(_x\) formation [23], which is reduced by ~150 K with the maximum water loading, WL\(_4\). However, this also causes a ~40% reduction in \(S_L\), with this drop in reactivity signifying reduced flame stability, particularly at the richest conditions.

A hybrid reactor network was developed to simulate the chemistry of the experimental swirling flame [10-11, 24]. Three inlets were used to provide fuel, air and steam flows, with three perfectly stirred reactors (PSR) to model the pre-mixing, flame and central recirculation zones (20% recycled flow [25]). The outlet from the flame zone fed a Plug Flow Reactor (PFR) to simulate reactions in the post-flame zone. The model was set up with representative combustor geometry, with residence times calculated from empirical flow conditions. Reactor temperature was defined from equilibrium conditions, with flame temperature.
calculated by the model, and set-points in the post-flame zone obtained from experimental results and system geometry. Heat loss was estimated to be 10%, with initial values compared to empirical results at baseline conditions to give favorable agreement with sampled concentrations. It should be emphasized that moderate uncertainty is associated with reactor networks, and the developed model was primarily used for empirical analysis, as opposed to prediction. Conditions were scaled with pressure, with all other geometric variables held constant, and only flow rates changed for each experimental set-point to give the plotted results. Plotted results from each modeled condition represent the combined NO + NO2 fractions, equivalent to the measurements taken with the analyzer described in section 2.2. Each input variable can be found alongside unmeasured additional concentrations predicted by the model in the supplementary material.
At baseline conditions of pressure and water loading the range of experimental Ø was broadened from 1 - 1.4, with sampled NO\textsubscript{x} concentrations plotted alongside modelled results in Fig. 3 (NH\textsubscript{3} data were only captured across the original specified range). Good agreement is noted between the sampled NO\textsubscript{x} concentrations and modelled predictions (both normalised to equivalent conditions), with slightly greater discrepancy for the NH\textsubscript{3} data. Low NO\textsubscript{x} concentrations achievable with rich mixtures have been shown by other researchers [12], but operation at such conditions incurs an efficiency drop due to unburned fuel. NO\textsubscript{x} reduction is achieved through a combination of reduced thermal formation with the Zeldovich mechanism [23] in the flame zone and NO consumption in the post-flame zone. Modelled inlet temperatures were increased to 515 K for the Ø=1.3 condition (giving a near equivalent A/F to that at Ø=1.2), with this thermal effect shown to approximately double the NO\textsubscript{x} product fraction, but still amounting to < 10% of the original Ø=1.2 case. The relative increase in NH\textsubscript{3} at richer Ø also leads to a significant increase in NH\textsubscript{2} in the post-flame zone, which enhances NO consumption through the chain-carrying reaction NH\textsubscript{2} + NO $\leftrightarrow$ NNH + OH and the terminating reaction NH\textsubscript{2} + NO $\leftrightarrow$ H\textsubscript{2}O + N\textsubscript{2}. The range of experimental Ø was restricted to Ø=1.2 - 1.3 for further experiments to focus on the configuration that generated favourable NO\textsubscript{x} concentrations (< 500 ppmv), without excess unburned H\textsubscript{2} and/or NH\textsubscript{3}.
The influence of moderately elevated pressure was explored in the specified range up to 0.185 MPa (P4), with operation at higher pressures requiring a fuel pre-heating vaporizer. Sampled NOx concentrations were nevertheless reduced by an order of magnitude across the experimental range, as shown in Fig. 4 (experimental markers are superimposed on the corresponding model prediction). This reduction is predicted to diminish as pressure is increased further, as demonstrated with modeled data at 0.4 MPa represented by the solid line, but suggests implementation at increased pressure could allow for leaner operation.

An increase in pressure was also predicted to give rise in unburned NH3 concentration, with experimental trends broadly agreeing, as demonstrated in Fig. 5. NH3 data could not be captured at P4 due to experimental run time limitations.

This influence of elevated pressure has recently been investigated numerically by Somarathne et al. [26] up to 0.5 MPa with swirling NH3/air flames, following work from Valera-Medina et al. [10] with CH4/NH3/air. The predicted reduction in NOx formation with pressure from the numerical NH3/air flame is small compared to the experimental results presented in Fig. 4. This was attributed [26] to the influence of OH in NH2 oxidation, as well as the role of HNO in NO production, and used to explain why results demonstrated a reduced influence at Ø=1.25. By contrast, a near equivalent fractional offset in NOx reduction is evident between the Fig. 4 Experimental (markers) and modeled (lines) NOx concentrations against Ø at elevated pressure.

In contrast, Fig. 5 Experimental (markers) and modeled (lines) NH3 concentrations against Ø at elevated pressure.
Model and experiments, across the range of $\phi$ for $\text{NH}_3/\text{H}_2/\text{air}$ flames presented here. Hayakawa et al. [27] demonstrated trends for reduced NO production with increased pressure, however the measured concentrations were significantly lower than those expected at $\phi=1$ when compared to this and other work [12]. This may have resulted from the removal of $\text{H}_2O$ from the sampled gas stream and the inability to stabilize a pilot flame—the influence of $\text{H}_2$ fraction was numerically investigated in this work, with an increase in $\text{H}_2/\text{NH}_3$ ratio shown to reduce the observed effect for NO reduction with increased pressure. To enhance the kinetic analysis further, Fig. 6 shows a comparison between the absolute rates of NO fractions produced from prominent reactions in both the flame zone P SR, and at a fixed axial distance (0.01 m) in the post-flame zone with changes in ambient pressure. The results identify an increase in reaction governing NO production as pressure rises in the flame zone (Fig. 6a). This is offset by near equivalent increases in NO consumption mechanisms, ultimately leading to a marginal reduction (~5%) in NO fraction in the flame zone. However, $\text{NH}_3$ and $\text{NH}_2$ become more significant as NO is consumed in the post-flame zone (Fig. 6b), as rates from the shown mechanisms increase with pressure. This results from enhanced OH consumption and the production of $\text{NH}_2$ from $\text{NH}_3 + \text{OH} \leftrightarrow \text{NH}_2 + \text{H}_2\text{O}$, which also leads to the formation of more $\text{NH}$ through chain carrying mechanisms with $\text{OH}$ and $\text{H}$. NO production rates also decrease from $\text{HNO} + \text{H} \leftrightarrow \text{NO} + \text{H}_2$, and whilst there is a relative increase through mechanisms such as $\text{NH} + \text{NO} \leftrightarrow \text{N}_2\text{O} + \text{H}$ and $\text{N} + \text{OH} \leftrightarrow \text{NO} + \text{H}$, fractions are diminished with overall NO consumption. Similar influences are evident at $\phi=1.3$, with a small reduction in NO formation in the flame zone, and enhanced consumption downstream, with the third body reaction $\text{NO} + \text{H} ( + \text{M}) \leftrightarrow \text{HNO} ( + \text{M})$ shown to be more prominent. The increase in unburned $\text{NH}_3$ primarily results from elevated $\text{NH}_2$ concentrations, with reactions $2\text{NH}_2 \leftrightarrow \text{NH}_3 + \text{NH}$ and $\text{NH}_2 + \text{H} + \text{M} \leftrightarrow \text{NH}_3 + \text{M}$; however, this influence is small compared to relative NO reduction.
Reactant Humidification is an effective process for controlling NO through the combined influences of reduced flame temperature, along with enhanced O consumption through the reaction $O + H_2 \rightarrow OH + OH$, to limit $N_2 + O \leftrightarrow NO + N$ [23]. In this work, the efficiency of employing humidification for NO reduction has been studied with fuel-bound nitrogen in $NH_3$, increasing $H_2O$ fraction to ~10% mol at $WL_4$. Beyond this concentration at $\phi = 1.3$ the flame began to appear unstable, flickering with intermittent detachment from the burner. Results are plotted against change in $\phi$ for each water loading at $P_i$ in Fig. 7, with experimental markers superimposed on each corresponding modeled line. Again, favorable agreement was observed between the model and experiments, with $H_2O$ loading shown to reduce product NO fraction by an order of magnitude across the specified range. Alongside the expected reduction in $N_2 + O \leftrightarrow NO + N$, temperature drop (~150 K at $WL_4$) in the flame zone inhibited NO production rate from the reaction $N + OH \leftrightarrow NO + H$, with Fig. 8 showing the water loading comparison for $\phi = 1.2$. Models also suggest a relative rise in NO consumption through $NH_2$ mechanisms, resulting from enhanced $NH_3$ decomposition, offset against an increase from $HNO + H \leftrightarrow NO + H_2$. Nevertheless, $H_2O$ addition ultimately leads to a greater relative decrease in flame zone NO fraction. Enhanced NO consumption in the post-flame zone (0.01 m) results from similar mechanisms to those shown for an increase in pressure, Fig. 7 Experimental (markers) and modeled (lines) NO concentrations against $\phi$ with increased humidification.

<table>
<thead>
<tr>
<th>Flame Zone</th>
<th>Post-Flame Zone</th>
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<tbody>
<tr>
<td>$NH_3NO \leftrightarrow NNH_{OH}$</td>
<td>$NH_3NO \leftrightarrow NNH_{OH}$</td>
</tr>
<tr>
<td>$HNOOH \leftrightarrow NOH_{2}$</td>
<td>$NH_3NO \leftrightarrow NNH_{OH}$</td>
</tr>
<tr>
<td>$NOH \leftrightarrow NOH_{2}$</td>
<td>$NH_3NO \leftrightarrow NNH_{OH}$</td>
</tr>
</tbody>
</table>

(a) $\phi = 1.2$

(b) $\phi = 1.2$

Absolute Rate of NO Production (frac)
reacting with \( \text{NH}_2 \) through the reactions discussed in section 4.1 (Fig. 8b), albeit by a reduced amount.

However, elevated \( \text{OH} \) concentrations (from \( \text{H}_2\text{O} \)) react to provide increased \( \text{NO} \) production through \( \text{N} + \text{OH} \leftrightarrow \text{NO} + \text{H} \), counteracting the enhanced consumption. Again, elevated \( \text{NH}_2 \) concentrations provide an increase in unburned \( \text{NH}_3 \), with values marginally higher than those measured from a rise in pressure (Fig. 5).

Due to differences in the mechanisms governing \( \text{NO}_x \) formation, the influences of pressure and \( \text{H}_2\text{O} \) can be combined to provide a greater overall reduction, as shown for \( \phi = 1.2 \) in Fig. 9 (similar trends were observed for \( \phi = 1.25 \) and 1.3) with measurements superimposed onto modelled data. The reduction in flame zone \( \text{NO} \) from \( \text{H}_2\text{O} \) addition is coupled with enhanced consumption at pressure to reduce production in the post-flame zone. It should be emphasised that whilst the combined influences of pressure and \( \text{H}_2\text{O} \) both cause an increase in unburned \( \text{NH}_3 \), when offset against a change in \( \phi \), the net effect is still a reduction (Fig. 5). Hence leaner operational \( \phi \) can be employed to reduce unburned fuel fractions without a \( \text{NO}_x \) penalty. This is particularly significant for gas turbine combustion, where the introduction of \( \text{H}_2\text{O} \) could also be achieved through an exhaust gas recirculation (EGR) configuration, to control operational combustor temperatures.
In an attempt to reduce unburnt NH$_3$ while retaining good NO$_x$ performance, the burner was reconfigured for staged combustion, with secondary air supplied around the quartz tube to form a downstream reaction zone. It was important to ensure the additional air flow did not dilute the primary flame, due to the demonstrated sensitivity of NO$_x$ to change in $\phi$. OH* chemiluminescence was used to validate the experimental technique, with Fig. 10 showing the difference between $\phi=1.2$ and $\phi=1.3$ without secondary air, along with an equivalent flame at SA$_3$.

Half-flames are shown (x = 0 mm on the burner center line) with the captured 200 image average (Fig. 10a) provided above Abel deconvoluted planar representations (Fig. 10b), and colour maps normalized to the equivalent intensity distribution. Minimal differences were observed, and integral intensity values ($I_{\text{OH}^*}$, following Runyon [19]) were obtained, giving an average difference of ~4% with all secondary air flows, compared to a measured increase of ~56% from $\phi=1.3$ to $\phi=1.2$.

Increasing secondary air flow to SA$_2$ reduced the global $\phi$ (calculated using combined combustion and secondary air) to 0.82 - 0.87 (change in primary flame denoted by $\phi_p=1.2$ - 1.3) to improve fuel burnout efficiency, where substantially higher fully premixed NO$_x$ concentrations would be expected [12]. Measured NO$_x$ values are plotted against change in water loading at $P_1$ (empty) and $P_3$ (shaded) in Fig. 11. A P SR reactor was insufficient for modeling this secondary flame with poor correlation obtained surprisingly, instead empirical trends for each experimental $P_1$ are superimposed. Contrasting tendencies are observed for a change in water loading, with a significant reduction observed at $\phi_p=1.2$, compared to a marginal increase at $\phi_p=1.3$. The images show OH* chemiluminescence for varying $\phi$ and secondary air flow.
It is suggested that this result from the influence of H\textsubscript{2}O on the change in primary NO\textsubscript{x} concentration, offset against an increase in unburned NH\textsubscript{3}. Minimal NO\textsubscript{x} produced in the primary flame at Ø\textsubscript{p} = 1.3 means formation mostly results from unburned NH\textsubscript{3} at the tube outlet, which increases with H\textsubscript{2}O loading. This explanation is supported by the relative increase also observed with rising pressure.

Measured NO\textsubscript{x} concentrations were also sensitive to secondary air flow rate, with the change in normalized values (open markers, dotted trend lines) from Ø\textsubscript{p} = 1.25 flame plotted against global Ø in Fig. 12. Measured NH\textsubscript{3} fractions (shaded) are also included with trend lines superimposed for clarity. It is apparent that an optimal point can be selected where both NO\textsubscript{x} and unburned NH\textsubscript{3} concentrations are minimized through careful choice of humidity, pressure, and secondary air flow. An operational point was specified at WL\textsubscript{2} and P\textsubscript{4}, with secondary air increased to give a global Ø of 0.98, generating respectable product NO\textsubscript{x} and NH\textsubscript{3} fractions of ~32 ppmvd (15% O\textsubscript{2}) and ~50 ppmvd (15% O\textsubscript{2}). The current burner design is simple, offering non-optimized mixing of the secondary air flow in particular. The facility is currently under development to enable enhanced mixture control with secondary humidity, and operation at significantly higher pressure.
Conclusions

The NO emissions produced by a premixed swirling NH\textsubscript{3}/H\textsubscript{2} flame are sensitive to operational Ø, increasing by several orders of magnitude across the experimental range. NO concentrations < 10 ppmv can be achieved at the richest conditions, however, this is accompanied by high fractions of unburned fuel and a corresponding reduction in combustion efficiency.

Consistent with recent model predictions for NH\textsubscript{3} flames, an increase in combustor pressure has been shown experimentally to lower exhaust NO\textsubscript{x} concentrations for the first time. Chemical kinetic models suggest this primarily results from enhanced NH\textsubscript{2} formation, acting to consume NO in the post-flame zone. Unburned NH\textsubscript{3} concentrations are also produced from reactive NH\textsubscript{2}, however, the influence is small compared to change in NO\textsubscript{x}.

Reactor humidification reduces NO\textsubscript{x} concentrations in the flame zone, limiting thermal production pathways with the extended Zeldovich mechanism. NO consumption in the post-flame zone is also enhanced through an increase in OH-produced NH\textsubscript{2}, and again results in higher unburned NH\textsubscript{3} concentrations. When offset against a change in Ø then net effect is still a reduction, meaning leaner operation can be employed to reduce unburned fuel fractions without incurring a NO\textsubscript{x} penalty.

The emissions performance of an NH\textsubscript{3}/H\textsubscript{2} swirling flame can be further improved by using a combination of humidity and secondary airflow in a staged configuration, and is enhanced by operation at elevated pressure. However, careful specification of equivalence ratio is required as excessive unburned NH\textsubscript{3} will eventually increase NO\textsubscript{x} formation in the secondary reaction zone.

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