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Influence of Variable Swirl on Emissions in a Non-Premixed Fuel-Flexible Burner at Elevated Ambient Conditions

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1 ABSTRACT

As alternative fuels are designated for future energy 33 2 applications, flexible combustor designs require considerable 3 34 4 development to ensure stable operation with reduced NOx 35 5 emissions. A non-premixed variable swirl burner was used to 36 experimentally appraise changes in NO production pathways, 6 37 with CH₄ NH₃, and H₂ flames, alongside intermediate fuel 7 38 blends. Maintaining an equivalent thermal power and flame 39 8 9 temperature between fuels, preheated reactants (500 K) were 40 supplied to the burner, with parametric changes made to 10 41 pressure $(1 - 6 bar_a)$ and swirl number (0.8 - 2.0). NO production 42 11 was characterized, alongside variations in flame structure and 43 12 topology, with a correlation demonstrated for exhaust emissions. 44 13 NO production was shown to be sensitive to combustor pressure, 45 14 providing an expected increase for CH_4 and H_2 flames. Emission 46 15 profiles from both NH_3 and H_2 flames are shown to be 16 47 17 significantly augmented by a change in swirl number. As NH₃ 48 fractions were increased in the H_2 blend, a decaying trend in NO 18 49 19 emissions was observed with an increase in pressure, and as a 50 function of mixture ratio. However, this behaviour was markedly 20 51 21 augmented by a change in swirl number and suggests that 52 further reductions may be possible at increased pressure. At the 22 53 low swirl/high pressure condition the NH₃/H₂ blend 23 54 outperformed pure H₂, providing lower NO concentrations. 24 55 Emissions data were normalised using the traditional dry/O_2 25 56 correction, alongside mass scaled by thermal power, with a 26 57 comparison provided. The corresponding differences in emission 27 58 formation pathways were investigated, alongside high-speed 28 59 29 OH* chemiluminescence to further elucidate findings. 60

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Keywords: Hydrogen, Combustion, Low-emission combustor,Turbulence, Fuel combustion.

NOMENCLATURE

A_{noz}	Area of the burner nozzle exit
A_{tan}	Tangential inlet area
AFT	Adiabatic flame temperature
\dot{m}_x	Mass flow rate of <i>x</i>
Р	Burner ambient pressure
q_{in}	Thermal input power
Q_{tan}	Tangential flow rate
Q_{total}	Total flow rate
r_{noz}	Burner nozzle radius
r_{tan}	Effective radius of the tangential inlet
Re	Reynolds number
Sg	Geometric swirl number
Т	Burner inlet temperature
\dot{V}_x	Volumetric flow of <i>x</i>
X_{x}	Mole fraction of <i>x</i>
ρ_x	Mass density of <i>x</i>
Φ	Global fuel-air equivalence ratio
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1. INTRODUCTION

Future energy transfer applications will require the use of alternative fuels to achieve evolving emissions targets, comprising a range of technologies to meet the differences between fluctuating renewable supply and transient demand. From the perspective of anthropogenic climate change, significant emissions from fossil fuels include carbon dioxide (CO₂) and methane (CH₄), alongside Nitrous oxide (N₂O), with the latter possessing a 100-year global warming of potential (GWP) ~265 times that of CO₂[1]. This is noteworthy, as even with the development and application of alternative, carbon-free fuels such as Hydrogen (H₂) and Ammonia (NH₃), the production of all emissions must be suitably quantified to facilitate the development of flexible, efficient, and non-

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polluting combustion systems. This includes more traditional 57 2 concern for formation of Nitrogen Oxides (NO_x, typically NO 3 and NO₂), which can also provide detrimental impacts on both -59 4 respiratory health and the environment. Whilst NO_x emissions 60 5 were already a primary consideration for natural gas fired 61 6 systems, this concern will continue to develop with the 62 7 challenging application of carbon-free alternative fuels. A range 63 8 of combustor configurations can be employed for optimized 64 9 emissions production, such as the lean premixed dry low 65 emission (DLE) strategy. However, fuel-flexible operation 10 66 11 remains a challenge, with associated stability issues such as 67 12 flashback [2]. Non-premixed combustors can offer advantages in 68 13 relation to flame stability, however often at the expense of 69 14 emissions performance. The aim of the research presented herein 70 is to appraise experimentally the relative emissions performance 15 71 16 of a fuel-flexible combustor at elevated conditions of 72 temperature and pressure, with changing burner geometry. 17

19 **1.1 Research Scope**

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20 The configuration employed comprised a turbulent fuel jet, 76 21 with co-annular swirling airflow, housed inside an optical 77 22 pressure casing. Three fuels (CH₄, H₂, and NH₃) were applied 78 23 both independently and in different mixture ratios, with the 79 24 influence of fuel-air turbulent mixing appraised using a variation 80 25 in geometric swirl number (defined in section 2.1) alongside an 81 26 increase in inlet ambient pressure. Numerous studies have 82 27 demonstrated the complex potential influence of varying swirl 83 28 number on emissions formation for both CH_4 [3, 4] and H_2 [5] 84 29 with non-premixed flames. Kim et al. [6] investigated the 85 30 influence of CH₄/H₂ blends and demonstrated an increase in local 86 31 temperature and NO_x with H₂ addition, offset by a reduction for 87 32 increasing swirl intensity. Results were also compared between 88 33 premixed and diffusion configurations, with the latter providing 89 34 lower emissions. Results from Gupta et al. [7] also suggested a 90 35 sensitivity for NO_x emissions to change with swirl in a premixed 91 36 CH₄ flame. Kashir et al. [8] demonstrated numerically a 92 37 reduction in flame length with both increasing swirl and H₂ 93 38 addition in a non-premixed CH4 flame, whereas De and Acharya 94 39 [9] showed greater swirl broadens the size of recirculation zone 95 40 for a fixed H₂ enriched mixture. The numerical work of Ilbas et 96 41 al. [10] predicted an increase in NO_x with swirl for H₂ enriched 97 42 fuels due to changing temperature gradients. The influence of 98 43 swirl strength has also been demonstrated in alternative 99 44 combustor architectures. Khalil and Gupta [11] investigated 100 45 swirl in a distributed CH₄ combustor and observed that higher 101 46 residence times and stronger swirl generates greater combustion 102 efficiency, whilst providing lower levels of NO and CO. Patel 103 47 48 and Shah [12] compared swirling and non-swirling inverse 104 49 diffusion flames and observed an increase in NO_x with H₂, and 105 50 more prominent in the case of non-swirling flow. 106

51 Experimental data are more limited in the context of fuel 107 52 switching for turbine relevant conditions, particularly for NH₃. 108 53 However, more data are available for trends observed with other 109 54 alternative fuels. Jalalatian [13] investigated the influence of 110 55 swirl and equivalence ratio on emissions for bluff-body 111 stabilized Propane (C3H8) flames and found a change in 56

sensitivity relative to Reynolds number with increasing swirl. Mansouri et al [14] saw a significant reduction in CO emissions with increased swirl from H₂ enriched C₃H₈ flames. Chiong et al [15] demonstrated a reduction in NO emissions with an increase in swirl in a biodiesel/natural gas fired GT combustor. Benaissa et al. [16] demonstrated numerically that increasing swirl number leads to improved mixing between air and fuel streams due to increasing the tangential flow velocity for biogas/H2 mixtures, with Anuj et al. [17] using simulations to show similarly that enhanced CH₄-air mixing with swirl number reduces peak temperature, and therefore NO_x.

In addition to turbulent mixing, ambient conditions are instrumental for defining the chemical kinetics of emissions formation, with contrasting trends demonstrated for different fuels – NO_x emissions from fuel blends comprising NH₃ have been shown to reduce with an increase in ambient combustor pressure [18-20]. This primarily results from augmented production of NO from OH, alongside consumption with NH and NH₂. This has been demonstrated for both premixed [18] and diffusion [20] flames, however the response is non-monotonic, as a function of NH₃-H₂ ratio. The change in stability limits and NO emissions from premixed swirling NH₃-air flames enriched with H₂/CH₄ were investigated at elevated pressure by Khateeb et al. [21]. Pressure rise was shown to widen the stability range whilst reducing NO emissions. The sensitivity to change in fuel ratio is explored in detail in this study, with the performance compared from a change in burner geometry.

Finally, Douglas et al. [22] recently quantified the potential for augmentation in emissions reporting, as a result of varying exhaust water fractions due to combustion of alternative fuels. Once dried, product NO_x concentrations were shown to be falsely inflated for H₂ blends compared with CH₄, making the traditional normalization process unsuitable for a direct comparison between fuels. In this study, product NO emissions were normalized using both traditional (dry ppmv at 15% oxygen (O₂), as is currently used in international standards - ISO 11042 [23]) and alternative methodologies proposed in contemporary research literature [22].

EXPERIMENTAL FACILITY AND DIAGNOSTICS 2.

This study was performed using a well-documented [18, 20] geometrically generic swirl burner designed and employed at Cardiff University's Gas Turbine Research Centre. The system has been employed previously in a range of configurations for the application of traditional [24, 25] and alternative fuels [26].

2.1 Pressurized Optical Combustor

The burner was employed in a non-premixed, co-annular flow configuration in this study. The assembly and pressure casing are presented schematically in cross-section in Fig. 1. The fuel injector comprises a 18 mm OD lance (Fig.1a), with a concentric 5 mm diameter plain-orifice for high velocity injection (Flow path 1) of the specified reactants. Mixtures were blended upstream of the injector from independent fuel supplies in a delivery manifold.

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FIGURE 1: CROSS-SECTIONAL SCHEMATIC OF THE BURNER AND CASING ASSEMBLY.

5 Compressed air entered the burner through the inlet plenum 6 (Fig.1b) with all fuel and air flows metered using a combination 7 of Coriolis mass-flow controllers ($\pm 0.35\%$). The plenum body 8 was preconditioned to the specified inlet temperature (T = 5009 K) using preheated air, dried to a dew point of -17°C. System 10 temperatures were allowed to stabilize for at least an hour before 11 data were captured. The premixing chamber (Fig.1c - unused in 12 this work) fed air to a radial-tangential swirler (Fig.1d) to 13 envelop the injected fuel flow (flow path 2), with a burner exit 14 nozzle radius equivalent to 20 mm. Both medium and high swirl nozzles were employed for this work (Fig. 2), with respective 15 geometric swirl numbers equivalent to Sg = -0.8 and Sg = -2.0, 16 17 as defined in Eqn. (1) [25]: 18

$$Sg = \frac{A_{noz} \cdot r_{tan}}{A_{tan} \cdot r_{noz}} \left(\frac{Q_{tan}}{Q_{total}}\right)^2 \tag{1)60}$$

20 where A_{noz} is the exit area of the burner nozzle, A_{tan} tangential 21 inlet area, r_{tan} the effective radius of the tangential inlet, r_{noz} the 22 nozzle radius, Q_{tan} is the tangential flow rate, and Q_{total} the total 23 flow rate.

24 Quartz windows (Fig.1e) facilitated optical access into the 25 insulated high-pressure casing (Fig.1g) with high-speed chemiluminescence measurements captured from the side, 26 27 perpendicular to the reactant flow direction. The flame was 67 28 housed within a cylindrical quartz confinement (Fig.1f) tube 68 29 with an expansion equivalent to 100 mm from the swirler nozzle. 69 30 The system was pressurized to each specified ambient condition 31 (P) using a water-cooled incremental back-pressure valve, 32 positioned downstream of the flame and temperature-33 conditioned emission sample probe. Further detail on the 34 experimental setup is provided in other studies [18, 19, 25], with 35 CAD models available on request.

Sg = 0.8 Sg = 2.0 2.85 mm 7.5 mm

FIGURE 2: COMPARSION OF SWIRLER GEOMETRIES EMPLOYED FOR THIS WORK.

2.2 Emissions Measurement

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Gaseous emissions were captured from the combustor exhaust, downstream of the quartz confinement using a 9-hole equal-area probe. The sample system was water-conditioned with a heat exchanger to regulate sample temperature to 433 K, alongside the pump, lines, and filter block following specifications in ISO-11042 [23].

NO, measurements were quantified hot/wet at 1 Hz using a heated vacuum chemiluminescence analyzer (Signal 4000VM). Additional flow was directed to a chiller, used to reduce the molar water concentration below 1% before downstream CO, CO₂ and O₂ measurements were undertaken using a combination of nondispersive infrared and paramagnetic analyzers (Signal instruments 9000MGA) respectively. Two methods of emissions normalization are compared in this work:

Normalization method 1: Dry ppm_V at 15% oxygen (O₂). Firstly, measured ppm_V concentrations (NOmeas) were corrected to equivalent dry values using Eqn. (2).

NO Dry =
$$\frac{\text{NOmeas}}{1 - X_{H_2O}}$$
 (2)

Exhaust water fractions (X_{H_2O}) were obtained from equilibrium modelling, with further detail provided in Section 3.1. Measured dry O_2 fractions (X_{O_2}) were then used to subsequently normalize readings to an equivalent reference 15% O_2 as shown in Eqn. (3) [23]

NO Dry 15%
$$O_2 = \text{NO Dry} \cdot \left(\frac{0.209 - 0.15}{0.209 - X_{O_2}}\right)$$
 (3)

Normalization method 2: Here, the mass of NO produced $(\dot{\mathbf{m}}_{NO})$ was scaled by the thermal power (q_{in}) supplied to the burner for each condition, calculated using Eqn. (4) below from Douglas et al. [22].

$$NO \frac{\dot{m}_{NO}}{q_{in}} = \frac{X_{NO} \ \rho_{NO} \ \dot{V}_{exhaust}}{\Delta h_c \ \rho_{fuel} \ \dot{V}_{fuel}} \tag{4}$$

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2 Measured NO concentrations were converted to equivalent 3 fractions (X_{NO}) . Volumetric fuel flow rate (\dot{V}_{fuel}) was simply calculated from the specified inlet conditions, along with 4 5 density. The unmeasured major components of the exhaust flow were determined using the equilibrium method as above for 6 X_{H_2O} , and converted to mass fractions. This was scaled by the 7 8 total mass flow through the system and converted for the 9 volumetric flow of the exhaust products. For mixtures, net heat 10 of combustion (Δh_c) was scaled by mass fraction.

experimental 11 After changing conditions, burner temperatures, pressures, flows, and emissions were stabilized 12 and held for a minimum of 120 measurements. Systematic 13 uncertainties comprising analyzer specification, linearization, 14 and span gas certification, were combined with any standard 15 deviations in measurement to give the total uncertainty 16 17 represented by the error bars shown in the plotted data. 18

19 2.3 Chemiluminescence

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20 High-speed OH* chemiluminescence imaging was 21 performed at each experimental condition to characterize 22 changes in flame topology. OH* measurements focused on the 23 well-known $A^{2}\Sigma + X^{2}\Pi$ OH* system [27]. Data were captured 24 using a combination of Phantom v1212 high-speed CMOS 25 camera, Specialised Imaging SIL40HG50 high-speed image 26 intensifier, UV lens (78 mm, f/11), and a narrow 315 nm (± 15 27 nm FWHM) bandpass filter. Further information on this specific 28 high-speed imaging setup is found in other works [19].

29 Chemiluminescence data were captured at 4 kHz, with the 30 image intensifier gated at 10 µs. A scaled target image gave the 31 image resolution, equal to ~5 pixels/mm, resulting in the 32 presented view field of 100 mm (axial y) by 50 mm (radial x). 33 Each chemiluminescence dataset was temporally averaged from 34 2000 instantaneous images and filtered using a 3 \times 3 pixel 35 median filter. The averaged images were then processed using a 36 modified Abel inversion algorithm, to provide a planar 37 representation of the three-dimensional, flame brush, as 38 employed previously [19, 26]. An axisymmetric comparison is 39 shown between the averaged raw OH* chemiluminescence (Fig. 40 3a) and equivalent Abel transform (Fig. 3b) for an example case 41 in Fig. 3 where the centerline of the burner nozzle is represented 42 by x = 50 mm and flow enters from the bottom. Due to space 43 limitations, only Abel deconvoluted half-flames are presented in 44 this paper, with the raw dataset available from the institutional 45 repository.

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3. EXPERIMENTAL SPECIFICATION 47

48 A comprehensive experimental matrix was specified for the 49 range of fuel mixtures, with only the salient results presented and discussed. The full experimental dataset of inlet conditions and 50 51 results is available through supplemental material and the 52 institutional repository. Experiments were performed using a 80 53 swirling diffusion flame with a fixed reactant inlet temperature 54 (T) of 500 ± 10 K. Air flows were specified to simulate a constant 55 turbine inlet temperature, and therefore varied for each fuel mixture. The CHEMKIN-PRO equilibrium reactor was used to 56

COLORMAP NORMALIZED TO IMAGE MAXIMUM



FIGURE 3: COMPARISON BETWEEN THE (a) TEMPORARILY AVERAGED RAW OH* CHEMILUMINESCENCE IMAGE AND (b) EQUIVALENT ABEL TRANSFORM.

generate adiabatic flame temperatures (AFT) for global fuel-air equivalence ratios (Φ) under conditions of constant enthalpy/pressure using the GRI-Mech 3.0 reaction mechanism [28] (53 chemical species and 325 reactions). Alternative mechanisms [29, 30] were appraised, with near negligible differences observed for AFT calculations (<0.4%). The baseline condition was defined for CH₄-air, with $\Phi = 0.6$, giving an equivalent AFT of ~1813 K. The corresponding Φ value for each fuel was then established to give the same approximate AFT, as shown in Table 1 alongside the range of experimental P and Sg. Precisely controlled experimental mass flow rates were captured and fed back into the equilibrium reactor to provide the range of simulated AFT values represented in Table 1 for each dataset. Differences in stoichiometric airflow requirements meant that even with changing Φ , air mass flow rates and bulk outlet velocities only varied by \pm 3% from the average value for all fuels at each equivalent ambient condition.

TABLE 1: SUMMARY OF EXPERIMENTAL CONDITIONS

Fuel (mol fraction)	P (MPa)	Sg	Φ	AFT (K)
1 CH ₄	0.11 - 0.6	0.8, 2.0	0.6	$1813 \pm \! 3$
1 H ₂	0.11 - 0.6	0.8, 2.0	0.503	$1808~{\pm}4$
$1 \text{ CH}_4 \longrightarrow \text{H}_2$ (0.2 incr.)	0.11	0.8, 2.0	0.6-0.503	1808 ±3
$0.2 \ CH_4, 0.8 \ H_2$	0.11 - 0.6	0.8	0.545	1807 ± 2
$0.25 \ NH_3 \ 0.75 \ H_2$	0.11 - 0.6	0.8, 2.0	0.546	1809 ±4
$0.15 \text{ NH}_3, 0.85 \text{ H}_2$	0.11 - 0.6	2.0	0.53	$1813 \ \pm 2$
$0.08 \text{ NH}_3, 0.92 \text{ H}_2$	0.11 - 0.6	2.0	0.52	1811 ± 3

CH₄/H₂ ratios were initially varied in fractional increments of 0.2, however small changes were observed until an equivalent H₂ fraction of 0.8 was reached. This blend was therefore specified for further detailed testing across the full range of P.

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Furthermore, after some preliminary investigation, three molar 1 39 2 NH₃/H₂ ratios were specified at 0.25/0.75, 0.15/0.85 and 40 3 0.08/0.92, as this was predicted to adequately capture the non-41 4 monotonic influence of pressure increase on NO production with 42 5 NH₃. Data were not captured for pure NH₃ flames in this work 43 6 as limitations in fuel vapor withdrawal meant equivalent ambient 44 7 combustor pressures could not be matched against the other 8 fuels. A new fuel delivery system will facilitate this in future 9 work.

10 To minimize differences in local velocities and combustor 11 residence time, mass flows, and therefore net thermal powers 12 were scaled with an increase in P at a ratio of 12.5 kW/0.11 MPa. This gave a maximum thermal power equivalent to ~68 kW at 13 the highest-pressure condition - 0.6 MPa. Hence, whilst nozzle 14 outlet velocities remained quasi-steady with an increase in P, 15 Reynolds numbers (Re) and therefore local turbulence intensity 16 increased in almost direct proportion. Taking the $\Phi = 0.6$ CH₄ 17 case as an example, at 0.11MPa the nozzle airflow Re was 18 19 ~8,500, increasing to ~46,200 at 0.6 MPa. The rise in ambient 20 pressure also mildly increased the AFT for each specified fuel 21 blend. This simulated change has been plotted for three example 22 fuel mixtures (CH₄-air, H₂-air, and NH₃/H₂-air) in Fig. 4, with a near equivalent offset of ~2 K for each mix across the 23 24 experimental range. The full range of calculated outlet velocities 25 and Reynolds numbers are provided for each experimental 26 condition in the supplemental material available through the 27 institutional repository.



28 FIGURE 4: CHANGE IN GLOBAL AFT WITH P FOR CH4 29 $(\Phi=0.6), H_2 (\Phi=0.503), AND 0.25 / 0.75 NH_3/H_2 (\Phi=0.548).$

31 **RESULTS AND DISCUSSION** 4.

33 4.1 CH₄ to H2

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34 To demonstrate the change in emissions production for a 35 variable fuel blend, H₂ fuel fraction - in CH₄- was increased in 36 increments of 0.2. Figure 5 presents a comparison between normalized NO concentrations processed using each 37 methodology outlined in Section 2.2 for this range in fuel 38

composition. Initially, only a moderate increase in NO is observed, increasing considerably once the H₂ fraction is increased above 0.6, consistent with other research findings [31, 32]. This is well-understood to be a result of increased peak temperatures for the H₂ enriched flames, leading to thermal NO production.



FIGURE 5: CHANGE IN NORMALIZED NO PRODUCTION FOR 46 INCREASING MOLAR H2 FRACTION WITH CH4 AT 0.11 MPa.

48 An increase in reactivity from H₂ enrichment typically acts 49 to shorten premixed flames, evidenced in previous studies [33]. 50 However, the inverse effect was demonstrated with the flame 51 configuration employed for this work, as shown in the Abel 52 transformed OH* chemiluminescence images presented in Fig. 53 6. Here, the flame brush elongates from the burner face and 54 taking OH* as a generalized marker for heat release [34], 55 provides an increase in flame zone residence time. This results 56 from a significant rise in the fuel injector jet velocity with H₂ 57 enrichment; as the combined effects of density change and 58 heating value are factored, the nozzle bulk jet velocity increases from $\sim 30 \text{ m} \cdot \text{s}^{-1}$ for CH₄ to $\sim 99.5 \text{ m} \cdot \text{s}^{-1}$ for H₂. This acts to reduce 59 the strength of the central recirculation zone (CRZ) formed by 60 the swirling airflow, as characterized for this swirler in previous 61 62 studies [24]. Nevertheless, the flame still appears stabilized in 63 the shear layer between the outward swirling bulk airflow and 64 CRZ, resulting in the familiar V-shape flame typically associated 65 with swirlers of this design [25, 33].

Whilst at first seeming subtle, the difference resulting from the change in normalization methodology provides a notable difference in NO emission performance. Production is shown to increase with a transition from CH_4 to H_2 by a factor of ~3.7 for the 15% dry O₂ case, versus ~2.7 from mass scaled by thermal power. This emphasizes the need to apply a suitable correction methodology to fully appraise burner performance when fuel switching.

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FIGURE 6: COMPARISON OF ABEL TRANSFORMED OH* CHEMILUMINESCENCE RESULTING FROM A FUEL SWITCH 4 FROM CH4 TO H2 AT 0.11 MPa.

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The comparative change in NO production between CH₄ 6 7 and H₂ was further evaluated by increasing ambient combustor 8 pressure. Figure 7 demonstrates this change across the full 9 experimental range from 0.11 to 0.6 MPa, with an intermediate 10 0.8/0.2_{mol} H₂/CH₄ fuel blend, alongside a comparison between the normalization methodologies for each fuel mixture. There is 11 12 near equivalent performance across the experimental range, with 13 NO production for both CH₄ and H₂ increasing by \sim 70% from 14 0.11-0.6 MPa regardless of which normalization methodology is 15 employed. Whilst H₂ initially shows a more prominent increase 16 at lower pressures, production begins to plateau, as observed in 17 other work [31], where typically NO_x emissions increase as a 18 general square root function with increasing pressure for non-19 premixed flames [31]. Applying a power law correlation to these 20 data, the pressure exponent increases marginally from the CH₄ 21 flame (0.308) to the H2 case (0.338), demonstrating an increased 22 sensitivity to pressure, as observed in [31].



FIGURE 7: CHANGE IN NORMALIZED NO PRODUCTION FOR 23 INCREASING AMBIENT COMBUSTOR PRESSURE, FOR CH4, H2 24 25 AND 0.8/0.2_{mol} H₂/CH₄ FUEL BLEND.

The 0.8/0.2_{mol} fuel blend, whilst producing lower overall emissions than the H₂ flame, demonstrates an increased sensitivity to pressure increase as NO emissions rise by over 115% at 0.6 MPa, yielding an equivalent pressure exponent of 0.458. The exponents presented were obtained using data normalized on a mass/power basis using method 2 - near equivalent exponents were observed if emissions are normalized using the traditional methodology. The 0.8/0.2_{mol} blend also shows minimal difference between the two emission correction methodologies, compared to the respective over/under correction given by using dry 15% dry O₂ for H₂/CH₄. Figure 8 shows the change in flame topology that results from an increase in pressure with Abel transformed OH* chemiluminescence images for the CH₄ and H₂ flames.

COLORMAP NORMALIZED TO IMAGE MAXIMUM



FIGURE 8: COMPARISON OF ABEL TRANSFORMED OH* CHEMILUMINESCENCE RESULTING FROM AN INCREASE IN COMBUSTOR PRESSURE FOR CH4 AND H2.

The CH₄ flame appears to elongate more substantially with an increase in pressure, and whilst the effect is observed for H₂ it is diminished in comparison. These trends are evident despite nozzle outlet velocities remaining quasi-constant between each condition, and near equivalent changes in Re between each fuel as pressure rises. This is attributed to a combination of change in momentum, mixing and heat release as pressure increases. As the flame elongates, this increases residence time in the flame zone, contributing to the enhanced thermal NO production, whilst post-flame NO_x production can also be exacerbated at increased pressure [35]. This is countered by the change in turbulent mixing that results from the change in density and Re. Tabet et al. [36] observed that the non-premixed H₂ flame

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reaction zone becomes thicker as pressure increases, with a fast 1 2 increase in peak temperature and from 1 to 5 atm reducing 3 thereafter and reducing thermal NO production. This contributes 4 to the plateau observed for H_2 in Fig, 7. At the highest 5 experimental pressure condition of 0.6 MPa the switch in fuel H₂ 6 from CH₄ results in an increase in NO production by near 7 equivalent factors of ~3.7 (15% dry O₂) and ~2.7 (mass/thermal 8 power) to the atmospheric case. However, prior to the plateau in 9 NO emissions from H₂, these factors increase to maximum values 10 of 4.1 (15% dry O₂) and 3.0 (mass/ thermal power) at 0.4 MPa.

4.2 Change in Swirl Number 12

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13 Corresponding experiments were performed with both CH₄ 14 and H_2 at high swirl conditions (Sg = 2.0), with Figure 9 15 highlighting the difference in measured NO between each 16 swirler for increasing pressure. Note, for clarity only data 17 normalized using method 2 for mass/thermal power are 18 presented.



19 FIGURE 9: CHANGE IN NORMALIZED NO PRODUCTION FOR 20 CH4, AND H2 WITH Sg AND INCREASING AMBIENT 21 COMBUSTOR PRESSURE. 22

23 There exists a marked contrast in response to changing swirl 24 number for each fuel: minimal differences in NO production are 25 observed for CH₄ at each pressure, whereas notable reductions 26 are evident for H₂ with increase in swirl number across the 27 experimental range (max 38%). Rashwan [3] demonstrated that 28 an increase in swirl number with CH₄ should enhance mixing, 29 thereby lowering peak temperatures as premixed behaviour is 30 approached. However, that effect was not observed for the CH₄ 31 flame in this work. Oh et al. [5] demonstrated that increase in 32 swirl vane angle improved mixing with an H₂ flame, reducing 33 flame length and pollutant NO_x emissions, with Kim et al. [6] 34 demonstrating equivalent trends. A comparison is made in Fig. 35 10 between the change in heat release and flame topology that 36 results from an increase in Sg with Abel transformed OH* 37 chemiluminescence images for both the CH₄ and H₂ flames.

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FIGURE 10: COMPARISON OF ABEL TRANSFORMED OH* CHEMILUMINESCENCE RESULTING FROM A CHANGE IN Sg, AT BOTH 0.11 AND 0.6 MPa FOR CH4 AND H2.

45 Similar trends are shown for each fuel at both elevated and 46 low pressure – the increase tangential momentum that results 47 from a higher Sg serves to strengthen the CRZ relative to the 48 injection of the central fuel jet. With the flame initially stabilized 49 in the shear layer, the overall flame length is shortened, drawing 50 downstream reacting flow from the combustor wall. It appears the enhanced H₂ diffusivity and reduction in chemical 52 timescales, supported by the improved mixing from increased 53 Sg, limits peak temperatures, therefore facilitating a drop in NO 54 production. Whilst overall NO concentrations for H₂ are lower 55 for Sg = 2.0 than 0.8, the increase that results from a rise in 56 combustor pressure is more pronounced, with the relative plateau 57 observed for Sg = 0.8 diminished with an equivalent pressure 58 exponent of 0.411, suggesting increased sensitivity. NO 59 concentrations almost double (compared to an increase of \sim 70%) 60 for this blend across the experimental range. Nevertheless, there 61 is still a marked improvement in the emissions produced from 62 the H₂ flame compared to CH₄, where the pressure exponent also 63 increased to 0.386, with NO only increasing by a factor of 1.7-64 2.3 (mass/ thermal power) for the pressure range considered.

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Contrasting behavior was observed once the burner was 1 -39 2 fueled with a 0.75/0.25_{mol} H₂/NH₃ blend. Figure 11 provides a 3 comparison in trends for NO emissions for each Sg with an 4 increase in ambient pressure across the specified range. Once 5 again, a comparison is made between each method of emissions 43 6 normalization, with a small offset between each case.

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7 FIGURE 11: CHANGE IN NORMALIZED NO PRODUCTION 8 FOR 0.75/0.25 H₂/NH₃ WITH Sg AND INCREASING AMBIENT 0 COMBUSTOR PRESSURE. 10

11 Non-premixed NH₃ and H₂/NH₃ flames have previously 12 been shown to generate a reduction in NO for an increase in pressure [18-20], however the profiles evident in Fig. 11 change 13 14 markedly with a switch in swirl number. For the Sg = 0.8 case, 15 NO quickly falls to values lower than measurements made with pure H_2 flames at P = 0.6 MPa. However, a relative plateau is 16 reached, and the emissions do not fall once P = 0.3 MPa is 17 exceeded. This behavior with increase in pressure has been 18 19 observed for premixed flames and is attributed to enhanced 20 consumption of NO with NH and NH2 alongside reduced 21 production from the reaction: HNO + OH \leftrightarrow NO + H₂O. A 22 reduction with increasing pressure is also observed for the Sg = 23 2.0 condition, however the inverse trend is evident, where the reduction in NO emissions appears to be increasingly enhanced 24 25 as pressure is increased. Figure 12 provides a comparison between Abel transformed OH* chemiluminescence for each 26 27 swirl number with this fuel blend. Once again, similar behavior 28 is evident for the CH₄ and H₂ flames - at Sg = 0.8, the flame is 29 stabilized along the shear layer, with a traditional V-shape, 30 similar to a premixed configuration. However, once Sg is 31 increased to 2.0, the strengthened CRZ appears to draw more 32 reacting flow from the combustor wall. A potential explanation for the observed trend in emissions is that for the high swirl case, 33 34 more reacting flow is being directed to where the flame is richest. 35 Pressure increase has been shown to provide an increase in NH₂ 36 production, which would act to consume NO formed in the shear layer. Recent work by Wang et al. [37] demonstrated that for a 37 38 premixed flame, using swirl number to increase residence time

reduced NO, N₂O and NO₂ emissions more efficiently than tripling the chamber's length. However, this may be partly attributed to a decrease in combustion efficiency. Interestingly, the study by Wang et al. [37] was performed at a fixed P = 0.2MPa, and at that single pressure the opposite trend is witnessed to that observed in this work - that is, markedly worse NO_x performance for the high swirl case (NO fractions more than doubled at Sg = 2.0), and convergence only observed at the highest pressure conditions. Regarding combustion efficiency, no marked reduction was evident throughout this work. A comparison between the NH₃/H₂, CH₄ and H₂ exhaust temperatures - measured using an R-type thermocouple positioned downstream of the quartz tube (Fig. 1f) - are shown in Fig. 13, alongside the measured differences in exhaust O2 for NH₃/H₂ across the pressure range. Whilst no substantial efficiency drop is evident, the potential exists for enhanced trace NH₃ slip, as would be expected with enhanced NH₂ production [18]. For this work NH₃ data could not be accurately measured, however it is not unreasonable to suggest that a small increase in NH₃ slip would result given the observed NO reductions, and previous work [18, 19, 37]. Additional research is required to evaluate how the trends observed would continue with further rise in combustor pressure. Nevertheless, results suggest that high swirl is favorable with non-premixed flames for fuels comprising NH₃ at significantly elevated pressure.



FIGURE 12: COMPARISON OF ABEL TRANSFORMED OH* 65 66 CHEMILUMINESCENCE RESULTING FROM A CHANGE IN Sg FOR 0.75/0.25_{mol} H₂/NH₃ ACROSS THE CHANGE IN P. 67



FIGURE 13: COMPARISON OF THE DIFFERENCES IN 1 2 MEASURED EXHAUST TEMPERATURES FOR NH₃/H₂, CH₄ AND 3 $H_2 AT Sg = 2.0$, WITH CHANGE IN O_2 FROM THE NH_3/H_2 FLAME.

5 4.3 change in NH₃/H₂ ratio

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Two additional ratios of H_2/NH_3 (0.85/0.15_{mol}, and 6 7 0.92/0.08_{mol}) were specified to characterize the sensitivity in 8 reducing NO with pressure. The aim was to identify the 9 approximate NH₃ concentration necessary for the beneficial 10 influence of increasing pressure to be realized. The change in 11 measured exhaust NO between each fuel blend is shown in Fig. 14. Again, data are presented normalized only using method 2 12 13 for mass/thermal power for clarity.



FIGURE 14: CHANGE IN NORMALIZED NO PRODUCTION 14 15 FOR THREE H₂/NH₃ BLENDS WITH INCREASING AMBIENT COMBUSTOR PRESSURE. 16

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18 Opposing trends were observed between each blend as 19 combustor pressure increased. The $0.92/0.08_{mol}$ H₂/NH₃ ratio 20 provided an increasing trend similar to the CH₄, H₂ and H₂/CH₄ 21 experiments, however with a reduced rate of increase. This

contrasts with the 0.75/0.25_{mol} H₂/NH₃ where a growing reduction was previously observed in Section 4.2. The 0.85/0.15_{mol} H₂/NH₃ mostly provides a relative plateau in NO, and marks approximately the ratio at which the beneficial impact of increasing pressure up to 0.4 MPa on NO production is achieved with this burner configuration. However, at the highest pressure condition (P = 0.6 MPa), a reduction in NO is evident for all applied fuel ratios, and presents a point of convergence for each mixture. This is noteworthy, as no distinct change in flame topology was evident, as shown in Fig. 15 with averaged Abel transformed OH* chemiluminescence.

COLORMAP NORMALIZED TO IMAGE MAXIMUM



34 FIGURE 15: COMPARISON OF ABEL TRANSFORMED OH* 35 CHEMILUMINESCENCE RESULTING FROM A CHANGE IN 36 H₂/NH₃ RATIO ACROSS THE CHANGE IN P. 37

The relative reduction in H₂ fraction provides a small increase in flame length at each pressure, with a more pronounced lengthening as the maximum pressure is achieved. However, the same overall flame shape is maintained, again with downstream reacting flow being drawn in from the combustor wall. There was an increase in bulk injector outlet velocity from ~92 to ~97 m·s⁻¹ as molar NH₃ fraction reduced from 0.25 to 0.08. However, this was near equivalent to the pure H_2 case, and considerably higher than the CH₄ condition. The chemiluminescence data suggests the enhanced recirculation resulting from high swirl is controlling the flow structure to give a near equivalent flame topology for all fuels.

50 Whilst the emissions convergence at P = 0.6 MPa requires further study, it should be noted that for the NH₃/H₂ blends used in this work, emissions performance observed at the highest pressure condition approaches that of the pure H₂ flame.

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Furthermore, at the Sg = 0.8 condition, better performance is 2 demonstrated for the 0.75/0.25_{mol} H₂/NH₃ mixture relative to H₂ 3 once P = 0.3 MPa is exceeded. The potential exists for these 4 trends to continue with a further increase in ambient combustor 5 pressure.

7 5. CONCLUSIONS

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8 To conclude, an experimental study was performed to 9 appraise the comparative emissions performance of a non-10 premixed, co-annular swirl burner supplied with CH₄ NH₃, and 11 H₂ in different mixture ratios. The influence of change in swirl 12 number and combustor ambient pressure were quantified.

13 A fuel switch from CH₄ to H₂ provided an increase in NO 14 production, with measured concentrations rising rapidly once 15 molar fractions of 0.6 were exceeded, consistent with previous 16 work. A rise in ambient combustor pressure leads to an increase 17 in NO production with both fuels and intermediate blends, and 18 attention must be given to the emissions normalization methodology adopted when appraising the relative performance 19 20 with a fuel switch.

21 An increase from medium to high radial-tangential swirl 22 (corresponding to geometric swirl numbers of 0.8 and 2.0) 23 provided no significant change in NO emissions production for 24 the CH₄ flame. However, significant reductions were observed for the non-premixed H₂ flame across all experimental 25 26 combustor pressures, reaching a maximum of 38%. Results are 27 discussed in relation to changes in flame topology, visualized 28 using high-speed OH* chemiluminescence.

29 A marked difference in NO production with increasing 30 pressure was observed if a molar H₂/NH₃ ratio of 0.75/0.25 is 31 employed. At medium swirl, NO concentrations drop rapidly and 32 reach a relative plateau that outperforms pure H₂ at the highest 33 pressure conditions. At high swirl, NO fractions continue to 34 decrease as combustor pressure is raised, with no observable 35 change in combustor efficiency across the evaluated range.

36 Finally, the molar H₂/NH₃ ratio was varied to investigate the 92 37 blend at which the beneficial impact of pressure increase on NO reduction is no longer realized. Results suggest this is near the 38 39 H₂/NH₃ ratio of 0.85/0.15_{mol}. Emissions performance converged 40as the maximum pressure investigated was approached, and a 41 decrease in NO was still measured for all fuel blends up to 0.6 42 MPa, beyond which further investigation is required. 43

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53 SUPPLEMENTARY MATERIAL

54 Supplementary material associated with this article can be 55 found, in the online version, at: https://www.cu-gtrc.co.uk/

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39 **Table Caption List**

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41 **TABLE 1:** Summary of experimental conditions

43 **Figure Caption List**

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46 FIGURE 1: Cross-sectional schematic of the burner and casing 47 assembly.

48 FIGURE 2: Comparison of swirler geometries employed for this 49 work

- 50 FIGURE 3: Comparison between the (a) temporarily averaged raw
- 51 OH* chemiluminescence image and (b) equivalent Abel transform.

52 **FIGURE 4:** Change in global AFT with P for CH₄ (Φ =0.6), H₂

 $(\Phi=0.503)$, and $0.25 / 0.75 \text{ NH}_3/\text{H}_2$ ($\Phi=0.548$). 53

FIGURE 5: Change in normalized NO production for increasing 54

55 molar H₂ fraction with CH₄ at 0.11 MPa. FIGURE 6: Comparison of Abel transformed OH* chemiluminescence resulting from a fuel switch from CH4 to H2 at 0.11 MPa.

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FIGURE 7: Change in normalized NO production for increasing ambient combustor pressure, for CH₄, H₂ and 0.8/0.2_{mol} H₂/CH₄ fuel blend.

FIGURE 8: OH* Comparison of Abel transformed chemiluminescence resulting from an increase in combustor pressure for CH4 and H2.

FIGURE 9: Change in normalized NO production for CH₄, and H₂ with Sg and increasing ambient combustor pressure.

FIGURE 10: Comparison of Abel transformed OH* chemiluminescence resulting from a change in Sg, at both 0.11 and 0.6 MPa for CH₄ and H₂. 70

FIGURE 11: Change in normalized NO production for 0.75/0.25 H₂/NH₃ with Sg and increasing ambient combustor pressure.

FIGURE 12: OH* Comparison of Abel transformed chemiluminescence resulting from a change in Sg for 0.75/0.25mol H₂/NH₃ across the change in P.

FIGURE 13: Comparison of the differences in measured exhaust temperatures for NH₃/H₂, CH₄ and H₂ at sg = 2.0, with change in O_2 from the NH₃/H₂ flame.

FIGURE 14: Change in normalized NO production for three

H₂/NH₃ blends with increasing ambient combustor pressure.

FIGURE 15: Comparison of Abel transformed OH* chemiluminescence resulting from a change in H₂/NH₃ ratio across the 82 change in P.