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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

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

30

31 Keywords: Hydrogen, Combustion, Low-emission combustor, 32 Turbulence, Fuel combustion.

33 NOMENCLATURE

52 1. INTRODUCTION

53 Future energy transfer applications will require the use of 54 alternative fuels to achieve evolving emissions targets, 55 comprising a range of technologies to meet the differences 56 between fluctuating renewable supply and transient demand. 57 From the perspective of anthropogenic climate change, 58 significant emissions from fossil fuels include carbon dioxide 59 (CO₂) and methane (CH₄), alongside Nitrous oxide (N₂O), with 60 the latter possessing a 100-year global warming of potential 61 (GWP) \sim 265 times that of CO₂ [1]. This is noteworthy, as even 62 with the development and application of alternative, carbon-free 63 fuels such as Hydrogen $(H₂)$ and Ammonia (NH₃), the 64 production of all emissions must be suitably quantified to 65 facilitate the development of flexible, efficient, and non-

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

19 1.1 Research Scope

18

20 The configuration employed comprised a turbulent fuel jet, 21 with co-annular swirling airflow, housed inside an optical 22 pressure casing. Three fuels $(CH_4, H_2,$ and $NH_3)$ were applied 23 both independently and in different mixture ratios, with the 24 influence of fuel-air turbulent mixing appraised using a variation 25 in geometric swirl number (defined in section 2.1) alongside an 26 increase in inlet ambient pressure. Numerous studies have 27 demonstrated the complex potential influence of varying swirl 28 number on emissions formation for both CH₄ [3, 4] and H₂ [5] number on emissions formation for both CH₄ [3, 4] and H₂ [5] 29 with non-premixed flames. Kim et al. [6] investigated the 30 influence of CH4/H2 blends and demonstrated an increase in local 31 temperature and NO_x with H_2 addition, offset by a reduction for 32 increasing swirl intensity. Results were also compared between 33 premixed and diffusion configurations, with the latter providing 34 lower emissions. Results from Gupta et al. [7] also suggested a 35 sensitivity for NO_x emissions to change with swirl in a premixed 36 CH4 flame. Kashir et al. [8] demonstrated numerically a 37 reduction in flame length with both increasing swirl and H² 38 addition in a non-premixed CH4 flame, whereas De and Acharya 39 [9] showed greater swirl broadens the size of recirculation zone 40 for a fixed H_2 enriched mixture. The numerical work of Ilbas et 41 al. [10] predicted an increase in NO_x with swirl for H_2 enriched 42 fuels due to changing temperature gradients. The influence of 43 swirl strength has also been demonstrated in alternative 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 47 efficiency, whilst providing lower levels of NO and CO. Patel 103 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. 95

51 Experimental data are more limited in the context of fuel 52 switching for turbine relevant conditions, particularly for NH₃. 108 53 However, more data are available for trends observed with other 54 alternative fuels. Jalalatian [13] investigated the influence of 110 55 swirl and equivalence ratio on emissions for bluff-body 56 stabilized Propane (C_3H_8) flames and found a change in

57 sensitivity relative to Reynolds number with increasing swirl. 58 Mansouri et al [14] saw a significant reduction in CO emissions 59 with increased swirl from H_2 enriched C_3H_8 flames. Chiong et al 60 [15] demonstrated a reduction in NO emissions with an increase 61 in swirl in a biodiesel/natural gas fired GT combustor. Benaissa 62 et al. [16] demonstrated numerically that increasing swirl 63 number leads to improved mixing between air and fuel streams 64 due to increasing the tangential flow velocity for biogas/H² 65 mixtures, with Anuj et al. [17] using simulations to show 66 similarly that enhanced CH4-air mixing with swirl number 67 reduces peak temperature, and therefore NO_x .

68 In addition to turbulent mixing, ambient conditions are 69 instrumental for defining the chemical kinetics of emissions 70 formation, with contrasting trends demonstrated for different 71 fuels – NO_x emissions from fuel blends comprising $NH₃$ have 72 been shown to reduce with an increase in ambient combustor 73 pressure [18-20]. This primarily results from augmented 74 production of NO from OH, alongside consumption with NH and 75 NH2. This has been demonstrated for both premixed [18] and 76 diffusion [20] flames, however the response is non-monotonic, 77 as a function of NH_3-H_2 ratio. The change in stability limits and 78 NO emissions from premixed swirling NH3-air flames enriched 79 with $H₂/CH₄$ were investigated at elevated pressure by Khateeb 80 et al. [21]. Pressure rise was shown to widen the stability range 81 whilst reducing NO emissions. The sensitivity to change in fuel 82 ratio is explored in detail in this study, with the performance 83 compared from a change in burner geometry.

84 Finally, Douglas et al. [22] recently quantified the potential 85 for augmentation in emissions reporting, as a result of varying 86 exhaust water fractions due to combustion of alternative fuels. 87 Once dried, product NO_x concentrations were shown to be 88 falsely inflated for H_2 blends compared with CH₄, making the 89 traditional normalization process unsuitable for a direct 90 comparison between fuels. In this study, product NO emissions 91 were normalized using both traditional (dry ppm v at 15% oxygen 92 (O2), as is currently used in international standards - ISO 11042 93 [23]) and alternative methodologies proposed in contemporary 94 research literature [22].

96 2. EXPERIMENTAL FACILITY AND DIAGNOSTICS

97 This study was performed using a well-documented [18, 20] 98 geometrically generic swirl burner designed and employed at 99 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 106 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.

FIGURE 1: CROSS-SECTIONAL SCHEMATIC OF THE 44
3 BURNER AND CASING ASSEMBLY. 3 BURNER AND CASING ASSEMBLY. 4

 $\frac{5}{6}$ Compressed air entered the burner through the inlet plenum $\frac{47}{48}$ 6 (Fig.1b) with all fuel and air flows metered using a combination $\frac{40}{49}$ 7 of Coriolis mass-flow controllers $(\pm 0.35\%)$. The plenum body $\frac{43}{50}$ 8 was preconditioned to the specified inlet temperature ($T = 500\frac{50}{51}$ 9 K) using preheated air, dried to a dew point of -17°C. System $\frac{51}{52}$ 10 temperatures were allowed to stabilize for at least an hour before $\frac{52}{53}$ 11 data were captured. The premixing chamber (Fig.1c - unused in $\frac{53}{54}$ 12 this work) fed air to a radial-tangential swirler (Fig.1d) to $\frac{54}{55}$ 13 envelop the injected fuel flow (flow path 2), with a burner exit $\frac{33}{56}$ 14 nozzle radius equivalent to 20 mm. Both medium and high swirl $\frac{50}{57}$ 15 nozzles were employed for this work (Fig. 2), with respective $\frac{37}{28}$ 16 geometric swirl numbers equivalent to Sg = \sim 0.8 and Sg = \sim 2.0, $\frac{38}{59}$ 17 as defined in Eqn. (1) [25]: 18

$$
Sg = \frac{A_{noz} \cdot r_{tan}}{A_{tan} \cdot r_{noz}} \left(\frac{Q_{tan}}{Q_{total}}\right)^2
$$
 (1)60
19
61
Exhaust water fraction

20 where A_{noz} is the exit area of the burner nozzle, A_{tan} tangential $\frac{62}{21}$ inlet area, r_{tan} the effective radius of the tangential inlet, r_{noz} the 21 inlet area, r_{tan} the effective radius of the tangential inlet, r_{noz} the ⁰⁵
22 nozzle radius, Q_{t} is the tangential flow rate, and Q_{t} the total ⁶⁴ 22 nozzle radius, Q_{tan} is the tangential flow rate, and Q_{total} the total 65
23 flow rate. flow rate.

24 Quartz windows (Fig.1e) facilitated optical access into the 66 25 insulated high-pressure casing (Fig.1g) with high-speed 26 chemiluminescence measurements captured from the side, 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 70 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.

37 FIGURE 2: COMPARSION OF SWIRLER GEOMETRIES
38 EMPLOYED FOR THIS WORK. 38 EMPLOYED FOR THIS WORK. 39

40 2.2 Emissions Measurement

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59

67

72

41 Gaseous emissions were captured from the combustor 42 exhaust, downstream of the quartz confinement using a 9-hole 43 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 46 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 52 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 (O_2) . Firstly, measured ppm_V concentrations (NOmeas) were corrected to equivalent dry values using Eqn. (2).

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

61 Exhaust water fractions (X_{H_2O}) were obtained from equilibrium modelling, with further detail provided in Section 63 3.1. Measured dry O_2 fractions (X_{O_2}) were then used to subsequently normalize readings to an equivalent reference 15% $O₂$ as shown in Eqn. (3) [23] 66

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

Normalization method 2: Here, the mass of NO produced (\dot{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
$$
\frac{\dot{m}_{NO}}{q_{in}} = \frac{X_{NO} \rho_{NO} \dot{V}_{exhaust}}{\Delta h_c \rho_{fuel} \dot{V}_{fuel}}
$$
 (4)

1 2 Measured NO concentrations were converted to equivalent 3 fractions (X_{NQ}) . Volumetric fuel flow rate (V_{fuel}) was simply 100 4 calculated from the specified inlet conditions, along with 5 density. The unmeasured major components of the exhaust flow 6 were determined using the equilibrium method as above for 7 X_{H_2O} , and converted to mass fractions. This was scaled by the 8 total mass flow through the system and converted for the 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.

11 After changing experimental conditions, burner 12 temperatures, pressures, flows, and emissions were stabilized 13 and held for a minimum of 120 measurements. Systematic 14 uncertainties comprising analyzer specification, linearization, 15 and span gas certification, were combined with any standard $\frac{5}{58}$ 16 deviations in measurement to give the total uncertainty $\frac{36}{59}$ 17 represented by the error bars shown in the plotted data. $\frac{60}{60}$ 18

18 2.3 Chemiluminescence $\begin{array}{cc} 61 & 61 \\ 19 & 2.3 & 61 \end{array}$

20 High-speed OH* chemiluminescence imaging was $\frac{62}{63}$ 21 performed at each experimental condition to characterize 64 22 changes in flame topology. OH^{*} measurements focused on the 65 23 well-known $A^2\Sigma + -X^2\Pi$ OH* system [27]. Data were captured 66 differences ob 23 well-known A-2+--X-11 Orthours 1212 high-speed CMOS $\frac{66}{67}$ 25 camera, Specialised Imaging SIL40HG50 high-speed image 68 26 intensifier, UV lens (78 mm, f/11), and a narrow 315 nm (\pm 15 $\frac{80}{69}$ 27 nm FWHM) bandpass filter. Further information on this specific $\frac{5}{70}$ 28 high-speed imaging setup is found in other works [19]. $\frac{1}{71}$

29 Chemiluminescence data were captured at 4 kHz, with the $\frac{1}{72}$ 30 image intensifier gated at 10 μs. A scaled target image gave the $\frac{7}{73}$ 31 image resolution, equal to \sim 5 pixels/mm, resulting in the $\frac{7}{74}$ 32 presented view field of 100 mm (axial y) by 50 mm (radial x). 7.5 33 Each chemiluminescence dataset was temporally averaged from $\frac{76}{76}$ 34 2000 instantaneous images and filtered using a 3 \times 3 pixel $\frac{70}{77}$ 35 median filter. The averaged images were then processed using a $\frac{7}{78}$ 36 modified Abel inversion algorithm, to provide a planar $\frac{70}{79}$ 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.

46 47 3. EXPERIMENTAL SPECIFICATION

48 A comprehensive experimental matrix was specified for the 49 range of fuel mixtures, with only the salient results presented and 50 discussed. The full experimental dataset of inlet conditions and 51 results is available through supplemental material and the 52 institutional repository. Experiments were performed using a 53 swirling diffusion flame with a fixed reactant inlet temperature 54 (T) of 500 \pm 10K. Air flows were specified to simulate a constant 82 55 turbine inlet temperature, and therefore varied for each fuel 56 mixture. The CHEMKIN-PRO equilibrium reactor was used to

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FIGURE 3: COMPARISON BETWEEN THE (a) TEMPORARILY 58 AVERAGED RAW OH* CHEMILUMINESCENCE IMAGE AND (b) 59 EQUIVALENT ABEL TRANSFORM. 60

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 ± 3
1 H ₂	$0.11 - 0.6$	0.8, 2.0	0.503	1808 ± 4
$1 CH4 \rightarrow H2$ $(0.2$ incr.)	0.11	0.8, 2.0	$0.6 - 0.503$	1808 ± 3
$0.2 \text{ CH}_4, 0.8 \text{ H}_2$	$0.11 - 0.6$	0.8	0.545	$1807 + 2$
$0.25 \text{ NH}_3 \, 0.75 \text{ H}_2$	$0.11 - 0.6$	0.8, 2.0	0.546	$1809 + 4$
0.15 NH_3 , 0.85 H_2	$0.11 - 0.6$	2.0	0.53	1813 ± 2
0.08 NH ₃ , 0.92 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.

78

1 Furthermore, after some preliminary investigation, three molar 2 NH3/H2 ratios were specified at 0.25/0.75, 0.15/0.85 and 3 0.08/0.92, as this was predicted to adequately capture the non-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 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.

13 This gave a maximum t 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. 13 This gave a maximum thermal power equivalent to ~68 kW at ≥ 120 14 the highest-pressure condition – 0.6 MPa. Hence, whilst nozzle 15 outlet velocities remained quasi-steady with an increase in P,
16. Beynolds numbers (Be) and therefore local turbulence intensity 16 Reynolds numbers (Re) and therefore local turbulence intensity 17 increased in almost direct proportion. Taking the $\Phi = 0.6 \text{ CH}_4$ 18 case as an example, at 0.11MPa the nozzle airflow Re was $\frac{9}{5}$ ₄₀ 19 \sim 8,500, increasing to \sim 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 23 near equivalent offset of \sim 2 K for each mix across the 24 experimental range. The full range of calculated outlet velocities 45 25 and Reynolds numbers are provided for each experimental $\frac{45}{46}$ 26 condition in the supplemental material available through the $\frac{40}{47}$ 27 institutional repository.

30 31 4. RESULTS AND DISCUSSION

32

33 4.1 CH₄ to H₂
34 To demons To demonstrate the change in emissions production for a $\frac{72}{73}$ 35 variable fuel blend, H_2 fuel fraction - in CH₄- was increased in 36 increments of 0.2. Figure 5 presents a comparison between 37 normalized NO concentrations processed using each 38 methodology outlined in Section 2.2 for this range in fuel

composition. Initially, only a moderate increase in NO is observed, increasing considerably once the H_2 fraction is 41 increased above 0.6, consistent with other research findings [31, 42 32]. This is well-understood to be a result of increased peak temperatures for the H_2 enriched flames, leading to thermal NO production.

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

47

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 53 6. Here, the flame brush elongates from the burner face and 54 taking OH^{*} as a generalized marker for heat release [34]. 54 taking OH* as a generalized marker for heat release [34],
55 provides an increase in flame zone residence time. This results provides an increase in flame zone residence time. This results 56 from a significant rise in the fuel injector jet velocity with H_2
57 enrichment: as the combined effects of density change and enrichment; as the combined effects of density change and 58 heating value are factored, the nozzle bulk jet velocity increases from ~30 m⋅s⁻¹ for CH₄ to ~99.5 m⋅s⁻¹ for H₂. This acts to reduce 60 the strength of the central recirculation zone (CRZ) formed by 61 the swirling airflow, as characterized for this swirler in previous 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 69 increase with a transition from CH₄ to H₂ by a factor of \sim 3.7 for 70 the 15% dry O_2 case, versus \sim 2.7 from mass scaled by thermal 71 power. This emphasizes the need to apply a suitable correction methodology to fully appraise burner performance when fuel switching.

 $\frac{1}{2}$ FIGURE 6: COMPARISON OF ABEL TRANSFORMED OH* 40
3 CHEMILUMINESCENCE RESULTING FROM A FUEL SWITCH 3 CHEMILUMINESCENCE RESULTING FROM A FUEL SWITCH
4 FROM CH4 TO H2 AT 0.11 MPa. FROM CH₄ TO H₂ AT 0.11 MPa.

5

6 The comparative change in NO production between CH4 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 CH₄ 11 the normalization methodologies for each fuel mixture. There is 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 H_2 initially shows a more prominent increase 100 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 $\rm H_2$ 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].

23 **FIGURE 7:** CHANGE IN NORMALIZED NO PRODUCTION FOR $\frac{55}{56}$ 24 INCREASING AMBIENT COMBUSTOR PRESSURE, FOR CH₄, H₂ $\frac{50}{57}$ 25 AND $0.8/0.2_{mol}H_2/CH_4$ FUEL BLEND.

27 The $0.8/0.2_{mol}$ fuel blend, whilst producing lower overall 28 emissions than the H2 flame, demonstrates an increased 29 sensitivity to pressure increase as NO emissions rise by over 30 115% at 0.6 MPa, yielding an equivalent pressure exponent of 31 0.458. The exponents presented were obtained using data 32 normalized on a mass/power basis using method 2 – near 33 equivalent exponents were observed if emissions are normalized 34 using the traditional methodology. The 0.8/0.2mol blend also 35 shows minimal difference between the two emission correction 36 methodologies, compared to the respective over/under correction 37 given by using dry 15% dry O_2 for H_2/CH_4 . Figure 8 shows the 38 change in flame topology that results from an increase in 39 pressure with Abel transformed OH* chemiluminescence pressure with Abel transformed OH* chemiluminescence images for the CH₄ and H₂ flames.

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41 FIGURE 8: COMPARISON OF ABEL TRANSFORMED OH* 42 CHEMILUMINESCENCE RESULTING FROM AN INCREASE IN
43 COMBUSTOR PRESSURE FOR CH4 AND H2 COMBUSTOR PRESSURE FOR CH₄ AND H₂.

45 The CH4 flame appears to elongate more substantially with 46 an increase in pressure, and whilst the effect is observed for H_2 47 it is diminished in comparison. These trends are evident despite 48 nozzle outlet velocities remaining quasi-constant between each 49 condition, and near equivalent changes in Re between each fuel 50 as pressure rises. This is attributed to a combination of change in 51 momentum, mixing and heat release as pressure increases. As 52 the flame elongates, this increases residence time in the flame 53 zone, contributing to the enhanced thermal NO production, 54 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 H_2 flame

GTP-23-1411 Pugh 6

1 reaction zone becomes thicker as pressure increases, with a fast 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 H_2 6 from CH4 results in an increase in NO production by near 7 equivalent factors of \sim 3.7 (15% dry O₂) and \sim 2.7 (mass/thermal 8 power) to the atmospheric case. However, prior to the plateau in 9 NO emissions from H_2 , these factors increase to maximum values $Sg = 0.8$ 10 of 4.1 (15% dry O2) and 3.0 (mass/ thermal power) at 0.4 MPa.

11

12 4.2 Change in Swirl Number

13 Corresponding experiments were performed with both CH4 14 and H_2 at high swirl conditions (Sg = 2.0), with Figure 9 100 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 $\frac{50}{51}$ 20 CH₄, AND H₂ WITH Sg AND INCREASING AMBIENT $\frac{52}{52}$
21 COMBUSTOR PRESSURE. COMBUSTOR PRESSURE. 22

22

23 There exists a marked contrast in response to changing swirl $\frac{54}{55}$ 24 number for each fuel: minimal differences in NO production are $\frac{55}{56}$ 25 observed for CH₄ at each pressure, whereas notable reductions $\frac{50}{57}$ 26 are evident for H₂ with increase in swirl number across the $\frac{57}{58}$ 27 experimental range (max 38%). Rashwan [3] demonstrated that $\frac{58}{59}$ 28 an increase in swirl number with CH₄ should enhance mixing, $\frac{60}{60}$ 29 thereby lowering peak temperatures as premixed behaviour is $\frac{00}{61}$ 30 approached. However, that effect was not observed for the CH₄ 62 31 flame in this work. Oh et al. [5] demonstrated that increase in $\frac{62}{63}$ 32 swirl vane angle improved mixing with an H_2 flame, reducing 64 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.

38 39 **COLORMAP NORMALIZED TO IMAGE MAXIMUM**

41 **FIGURE 10:** COMPARISON OF ABEL TRANSFORMED OH^{*}
42 CHEMILUMINESCENCE RESULTING FROM A CHANGE IN Sg. CHEMILUMINESCENCE RESULTING FROM A CHANGE IN Sg, 43 AT BOTH 0.11 AND 0.6 MPa FOR CH4 AND H2.

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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 H_2 diffusivity and reduction in chemical timescales, supported by the improved mixing from increased 53 Sg, limits peak temperatures, therefore facilitating a drop in NO production. Whilst overall NO concentrations for H_2 are lower 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 observed for $Sg = 0.8$ diminished with an equivalent pressure exponent of 0.411, suggesting increased sensitivity. NO concentrations almost double (compared to an increase of \sim 70%) for this blend across the experimental range. Nevertheless, there is still a marked improvement in the emissions produced from the H_2 flame compared to CH₄, where the pressure exponent also increased to 0.386, with NO only increasing by a factor of 1.7-2.3 (mass/ thermal power) for the pressure range considered.

1 Contrasting behavior was observed once the burner was 39 2 fueled with a $0.75/0.25$ _{mol} H₂/NH₃ blend. Figure 11 provides a 40 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 6 normalization, with a small offset between each case.

7 FIGURE 11: CHANGE IN NORMALIZED NO PRODUCTION 63 8 FOR $0.75/0.25$ H₂/NH₃ WITH Sg AND INCREASING AMBIENT Ω COMBIENT BEESSURE COMBUSTOR PRESSURE.

11 Non-premixed NH₃ and H₂/NH₃ flames have previously 12 been shown to generate a reduction in NO for an increase in 13 pressure [18-20], however the profiles evident in Fig. 11 change 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
 $16 \text{ mm} \cdot \text{H} = 0.6 \text{ MP} \cdot \text{H} = 0.6 \$ 16 pure H₂ flames at P = 0.6 MPa. However, a relative plateau is 17 reached, and the emissions do not fall once $P = 0.3$ MPa is 18 exceeded. This behavior with increase in pressure has been 19 observed for premixed flames and is attributed to enhanced 20 consumption of NO with NH and NH_2 alongside reduced 100 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 24 reduction in NO emissions appears to be increasingly enhanced 25 as pressure is increased. Figure 12 provides a comparison $Sg = 2.0$ 26 between Abel transformed OH* chemiluminescence for each 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 0 (mm) 32 reacting flow from the combustor wall. A potential explanation 33 for the observed trend in emissions is that for the high swirl case, 65 34 more reacting flow is being directed to where the flame is richest. 65 35 Pressure increase has been shown to provide an increase in NH2 36 production, which would act to consume NO formed in the shear 37 layer. Recent work by Wang et al. [37] demonstrated that for a 38 premixed flame, using swirl number to increase residence time

reduced NO, N_2O and NO_2 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 $P = 0.2$ MPa, and at that single pressure the opposite trend is witnessed to that observed in this work – that is, markedly worse NO_x 45 performance for the high swirl case (NO fractions more than 46 doubled at $S_g = 2.0$, and convergence only observed at the 47 highest pressure conditions. Regarding combustion efficiency, 48 no marked reduction was evident throughout this work. A 49 comparison between the $NH₃/H₂$, CH₄ and H₂ exhaust 50 temperatures – measured using an R-type thermocouple 51 positioned downstream of the quartz tube (Fig. $1f$) – are shown 52 in Fig. 13, alongside the measured differences in exhaust O_2 for 53 NH₃/H₂ across the pressure range. Whilst no substantial 53 $NH₃/H₂$ across the pressure range. Whilst no substantial 54 efficiency drop is evident, the potential exists for enhanced trace 54 efficiency drop is evident, the potential exists for enhanced trace 55 NH₃ slip, as would be expected with enhanced $NH₂$ production 56 [18]. For this work $NH₃$ data could not be accurately measured, 57 however it is not unreasonable to suggest that a small increase in however it is not unreasonable to suggest that a small increase in 58 $NH₃$ slip would result given the observed NO reductions, and 59 previous work [18, 19, 37]. Additional research is required to previous work [18, 19, 37]. Additional research is required to 60 evaluate how the trends observed would continue with further 61 rise in combustor pressure. Nevertheless, results suggest that 62 high swirl is favorable with non-premixed flames for fuels comprising $NH₃$ at significantly elevated pressure.

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

GTP-23-1411 Pugh 8

1 **FIGURE 13:** COMPARISON OF THE DIFFERENCES IN
2 MEASURED EXHAUST TEMPERATURES FOR NH/H2 CH4 AND 0.11MPa 2 MEASURED EXHAUST TEMPERATURES FOR NH3/H2, CH4 AND 3 H₂ AT Sg = 2.0, WITH CHANGE IN O₂ FROM THE NH₃/H₂ FLAME.

5 4.3 change in NH₃/H₂ ratio

4

6 Two additional ratios of H_2/NH_3 $(0.85/0.15_{mol})$, and
7, 0.02/0.08 at an approximate the approximation of the constitutive in 7 0.92/0.08_{mol}) were specified to characterize the sensitivity in 8 reducing NO with pressure. The aim was to identify the 8 reducing NO with pressure. The aim was to identify the 9 approximate NH₂ concentration necessary for the beneficial 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. 12 14. Again, data are presented normalized only using method 2 13 for mass/thermal power for clarity.

14 **FIGURE 14:** CHANGE IN NORMALIZED NO PRODUCTION 47 chemilumines 15 FOR THREE H_2/NH_3 BLENDS WITH INCREASING AMBIENT 48 16 COMBUSTOR PRESSURE.
17

17 17
18 Opposing trends were observed between each blend as $\frac{50}{51}$ 19 combustor pressure increased. The $0.92/0.08_{\text{mol}}$ H₂/NH₃ ratio $\frac{51}{52}$ 20 provided an increasing trend similar to the CH₄, H₂ and H₂/CH₄ $\frac{52}{53}$ 21 experiments, however with a reduced rate of increase. This

22 contrasts with the $0.75/0.25$ _{mol} $H₂/NH₃$ where a growing 23 reduction was previously observed in Section 4.2. The 24 $0.85/0.15_{mol}$ H₂/NH₃ mostly provides a relative plateau in NO, 25 and marks approximately the ratio at which the beneficial impact 26 of increasing pressure up to 0.4 MPa on NO production is 27 achieved with this burner configuration. However, at the highest 28 pressure condition ($P = 0.6$ MPa), a reduction in NO is evident 29 for all applied fuel ratios, and presents a point of convergence 30 for each mixture. This is noteworthy, as no distinct change in 31 flame topology was evident, as shown in Fig. 15 with averaged 32 Abel transformed OH* chemiluminescence.

COLORMAP NORMALIZED TO IMAGE MAXIMUM

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

38 The relative reduction in H₂ fraction provides a small 39 increase in flame length at each pressure, with a more 40 pronounced lengthening as the maximum pressure is achieved. 41 However, the same overall flame shape is maintained, again with 42 downstream reacting flow being drawn in from the combustor 43 wall. There was an increase in bulk injector outlet velocity from 44 ~92 to ~97 m⋅s⁻¹ as molar NH₃ fraction reduced from 0.25 to 45 0.08. However, this was near equivalent to the pure H_2 case, and 46 considerably higher than the CH4 condition. The chemiluminescence data suggests the enhanced recirculation resulting from high swirl is controlling the flow structure to give 49 a near equivalent flame topology for all fuels.

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 H_2 flame.

1 Furthermore, at the $Sg = 0.8$ condition, better performance is 57 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

6

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 19 methodology adopted when appraising the relative performance 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 25 for the non-premixed H_2 flame across all experimental 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 H_2 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 37 blend at which the beneficial impact of pressure increase on NO 38 reduction is no longer realized. Results suggest this is near the 39 H₂/NH₃ ratio of 0.85/0.15_{mol}. Emissions performance converged 40 as 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/ 56

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36

37 38

39 Table Caption List

40

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

42 43 Figure Caption List

- 44
- 45

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
- 53 (Φ =0.503), and 0.25 / 0.75 NH₃/H₂ (Φ =0.548).

54 FIGURE 5: Change in normalized NO production for increasing

55 molar H2 fraction with CH4 at 0.11 MPa.

56 FIGURE 6: Comparison of Abel transformed OH* 57 chemiluminescence resulting from a fuel switch from CH₄ to H₂ at 0.11
58 MPa. MP_a

59 FIGURE 7: Change in normalized NO production for increasing 60 ambient combustor pressure, for CH₄, H₂ and $0.8/0.2_{\text{mol}}$ H₂/CH₄ fuel 61 blend.

62 FIGURE 8: Comparison of Abel transformed OH* 63 chemiluminescence resulting from an increase in combustor pressure 64 for CH4 and H2.

65 FIGURE 9: Change in normalized NO production for CH4, and H₂ 66 with Sg and increasing ambient combustor pressure.

67 FIGURE 10: Comparison of Abel transformed OH* 68 chemiluminescence resulting from a change in Sg, at both 0.11 and 0.6 69 MPa for CH4 and H2.

70 FIGURE 11: Change in normalized NO production for 0.75/0.25 71 H2/NH³ with Sg and increasing ambient combustor pressure.

72 FIGURE 12: Comparison of Abel transformed OH* 73 chemiluminescence resulting from a change in Sg for $0.75/0.25$ mol
74 H₂/NH₃ across the change in P H₂/NH₃ across the change in P.

75 FIGURE 13: Comparison of the differences in measured exhaust 76 temperatures for NH₃/H₂, CH₄ and H₂ at sg = 2.0, with change in O_2
77 from the NH₃/H₂ flame. from the $NH₃/H₂$ flame.

78 FIGURE 14: Change in normalized NO production for three

79 H2/NH3 blends with increasing ambient combustor pressure.

80 FIGURE 15: Comparison of Abel transformed OH* 81 chemiluminescence resulting from a change in H2/NH3 ratio across the 82 change in P.