Experimental study on the effect of pressure on single and two stage combustion of decomposed ammonia (NH$_3$–H$_2$–N$_2$) blends over a swirl stabilized burner

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ABSTRACT

Ammonia (NH$_3$) is a practical hydrogen vector for gas turbine power generation, but struggles to be deployed commercially due to many unresolved technical and fundamental challenges. This experimental investigation focuses on combustion of fuel blends representatives of various NH$_3$ decomposition (or cracking) ratios in a combustor fitted with a swirl-stabilized burner, exploring pressures ranging from atmospheric to 10 bar. The study presents measurements of flame stability and emissions of NOx and N$_2$O in combustor strategies relevant to both conventional single lean operation and staged with a rich primary zone, as a function of decomposition ratios (DR) and primary zone equivalence ratio. The findings reveal a notable pressure-dependency of nitrogen oxides (NOx) during combustion of ammonia containing fuels. For a fully decomposed mixture (H$_2$–N$_2$ at 75/25 %vol.), NOx emissions increase with increasing pressure. Conversely, when ammonia is present in the fuel, the trend reverses. However, the majority of NOx reduction occurs within a limited range of pressure increase and reaches a plateau thereafter. The transition at which the pressure effect declines depends on other factors, and was seen to occur between 5 and 9bar in the conditions of this study. Notably, NOx levels in flames containing NH$_3$ are significantly higher, rendering the pressure sensitivity on NOx primarily governed by NH$_3$ presence, irrespective of its concentration in the fuel (i.e. DR). Furthermore, the study reveals a non-monotonic relationship between NOx emissions and NH$_3$ concentration in the fuel, peaking at mid-decomposition ratios. All factors tending to weaken flame stability (e.g. increasing fuel NH$_3$ content, approaching lean/rich blow off limits, or increasing burner exit velocity) favour emissions of N$_2$O emissions. As a general trend, it appears that longer residence times, increased pressure, and rich primary zone operation are all parameters limiting NOx formation from all partially decomposed blends of ammonia, but that they only positively add to a certain limit of a few hundred ppm. This NOx plateau level being higher at higher NH$_3$ content in the fuel. Finally, the fuel obtained by perfectly decomposing ammonia appears to be the optimal fuel associating strong flame stability and low NOx propensity, even for the non fully premixed burner type of this study.

Novelty and Significance Statement: This research compiles experimental data of NH$_3$–H$_2$–N$_2$ blends under high pressure conditions, which are the first available in the range 5 to 10 bar. Experimental evidence of the beneficial effect of pressure on NOX has been shown earlier, but these are the first showing that this effect flattens out beyond 5 bar and combines with other operating factors, being therefore a valuable finding for assessing the potential of NOx emissions in gas turbine conditions. The study covers combustion modes in single and 2 stage (with rich primary zone) configurations and a variety of parametric variations. The study assesses the whole ammonia decomposition (cracking) range and highlights the combined effects of decomposition ratio and the other parameters investigated. Although there are many numerical studies on fundamental configurations with focus on kinetics with ammonia (flat flame, plug flow reactors, etc.), our results are obtained with a realistic burner configuration (swirl stabilized) at up to 100 kW power, giving the study an industrial relevance.

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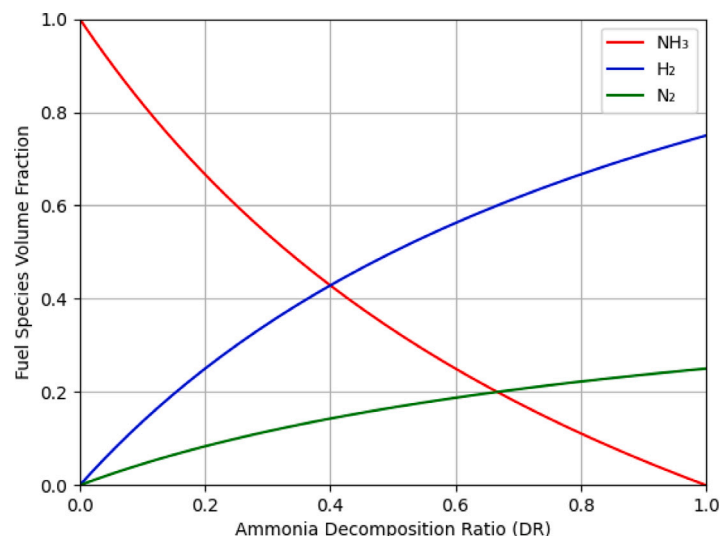*E-mail address:* mario.ditaranto@sintef.no (M. Ditaranto).

Fig. 1. Fuel blend composition as a function of decomposition (cracking) ratio (DR).

1. Introduction

Hydrogen, a carbon-free fuel, poses challenges in transportation and storage due to its physical properties. One promising solution involves converting hydrogen into ammonia, a liquid that is widely distributed globally through pipelines, rail, road, and ship networks. While the conventional Haber-Bosch process for ammonia production is known, it incurs additional costs in the already expensive hydrogen production. Recent interest in using ammonia as a fuel has sparked extensive research, particularly for power and heat generation. Ammonia, available in liquid or gas form, is suitable for gas turbines known for their high electrical efficiency and power density (size per power generated). However, direct use of ammonia as fuel presents challenges in stability and NOx emissions during combustion [1]. An alternative approach involves partial decomposition (also referred to as cracking) of ammonia, creating a fuel mixture composed of NH_3-H_2-N_2 in the proportion shown in Fig. 1, which has far better combustion properties, but at the expense of an endothermal reaction. This paper focuses on the practical application of partially decomposed ammonia blends in gas turbines operating under elevated pressure conditions.

A respectable amount of studies has focused on the effect of pressure on two fundamental properties of ammonia combustion: the laminar flame speed and the ignition delay time. It is shown experimentally and reproduced numerically with most of the kinetic mechanisms, that increasing pressure leads to a decrease of NH_3 containing flame speed [2] as is often the case for most fuels. Mei et al. [3] also noted that as the decomposition ratio increases, the pressure sensitivity gets stronger due to the increase of H_2 in the fuel mixture, making the reaction $H + O_2 (+ M) = HO_2 (+ M)$ more important. However, due to a sub-unity Lewis number and strong differential diffusion effects, the turbulent flame speed increases at lean conditions with pressure via increase in turbulent Reynolds number [4,5]. Several studies showed that the ignition delay time decreases with an increase in pressure, with up to a reported factor of around 7 between 1.4 and 11 atm at 2000 K in Mathieu and Petersen [6]. These properties are often used as validation for kinetic models adapted for ammonia as fuel, with particular focus on the formation and destruction steps of NO. A review of validated kinetic mechanisms in various conditions can be found in Valera-Medina et al. [7].

While these studies mainly utilized fundamental reactor systems and flames such as constant volume bomb, laminar flat flame, etc., limited research addresses the effect of pressure on realistic flames found in industrial boilers and engines which are characterized by complex tur-bulent mixing of flows and premixed/non-premixed chemistry. Table 1 lists the only experimental studies, to the knowledge of the authors, which are based on applied burner flames of ammonia containing fuels under pressurized conditions. Some of these have actually been realized in real gas turbine conditions [8–11], although small size ones with moderate pressure ratio. In the pioneering work done in Japan [9,11], several combustor modifications and fuel mixtures were tested and the staged rich-lean mode of operation was introduced. In Kurata et al. [10] it was achieved NOx emissions of 337 ppm @16%O_2 in the engine with a pressured ratio of 3. Other recent experiments at realistic gas turbine conditions have been reported in a 100 kWe micro gas turbine with a combustor pressure of 4.2 bar [8]. It was shown possible to reach 63% NH_3 in CH_4 provided that the pilot flame power was increased and with a loss of combustion efficiency. High levels of both NOx (1500–2200 ppm) and N_2O (100–1000 ppm) were recorded, to the point of cancelling out the benefit of using a decarbonized fuel due to the very strong global warming potential of N_2O. With the same burner on a laboratory test stand at 4.5 bar, up to 70% NH_3 could be added to methane to achieve flame lean stability behaviour similar to that of the methane flame [12], with high N_2O when approaching lean blow off limit. These two studies highlight the challenge to transfer laboratory performance to an actual engine. The largest scale at which ammonia has been used as fuel in a gas turbine have been performed by IHI in a 2 MW engine by co-firing natural gas with up to 20%LHV ammonia [13]. NOx from rig tests at atmospheric pressure and lean conditions were seen to be reduced from ca. 450 ppm @16%O_2 to 300 in the engine. In two stage combustion mode, the engine achieved 230 ppm @16%O_2 with a diffusion burner and a primary zone equivalence ratio of 0.85. Nevertheless, due to the stiffness of real engine studies, the pressure could not be varied in a parametric manner in any of these studies.

Pressure has been reported to affect ammonia containing flame stability and in that sense, to play a role on fundamental properties of flame propagation. In Okafor et al. [14] pressure contributed to extend the fuel rich stability limit, attributed to the increase of fuel entrainment in the recirculation zones which are responsible for flame stabilization. They focused the analysis on combined pressure and wall heat losses effects and suggested that the latter could have a contradicting and dominating effect on NOx behaviour when pressure increases. Similarly, Khateeb et al. [15] could extend the flashback limit to higher NH_3 contents in the fuel on a swirl stabilized full premixed burner characterized by low bulk velocity, thanks to an increase in pressure up to 5 bar. The effect of pressure was measured in the range 1 to 4 bar and showed that it was most effective in conditions were NOx were highest. Competitively low NOx was obtained in mixture with H_2

Table 1

Experimental studies on high pressure combustion of ammonia containing fuels with application relevant burners.

Reference	Year	Burner type	Fuel	P_{max} (bar)
This study [–]	2024	Rapid mixing	NH_3-H_2-N_2	10
Ditaranto et al. [16]	2023	Non-premixed	NH_3-CH_4	8
Pugh et al. [17]	2023	Non-premixed	NH_3-H_2	6
Ávila et al. [8]	2023	Piloted premixed	NH_3-CH_4	4.2
Wang et al. [18]	2023	Perfectly premixed	NH_3-CH_4	2
Avila et al. [12]	2022	Piloted premixed	NH_3-CH_4	4.5
Ditaranto et al. [19]	2022	Industrial DLE	NH_3-CH_4	6
Khateeb et al. [15]	2021	Perfectly premixed	NH_3-CH_4; NH_3-H_2-N_2	5
Ditaranto et al. [20]	2021	Industrial DLE	NH_3-H_2-N_2	4
Okafor et al. [14]	2021	Perfectly premixed	NH_3-CH_4	5
Kurata et al. [10]	2019	Premixed + non-premixed	NH_3-CH_4	3
Iki et al. [11]	2017	Premixed + non-premixed	NH_3-Kerosen	3
Kurata et al. [9]	2017	Premixed + non-premixed	NH_3-CH_4	3
Pugh et al. [21]	2019	Perfectly premixed	NH_3-CH_4; NH_3-H_2	1.8
Pugh et al. [22]	2020	Perfectly premixed	NH_3-CH_4; NH_3-H_2	1.7
Valera-Medina et al. [23]	2017	Perfectly premixed	NH_3-CH_4	2

at very lean conditions, however it is suspected that N_2O levels were excessive (N_2O and NH_3 were not measured).

By using the staged combustion strategy proposed in Somarathne et al. [24], ammonia can be partially burned and decomposed to hydrogen in oxygen poor atmosphere, limiting conversion to NOx through the fuel-NO route Then the secondary stage completes the combustion of hydrogen at a much lower NO production rate. The experimental study reported in Ditaranto et al. [20] confirmed the strategy with all blends corresponding to various degrees of decomposed ammonia, but showed that for medium to high decomposition ratios, the increase in adiabatic flame temperature resulting from a higher concentration of hydrogen in the fuel, reduces the positive effect of a rich primary zone (PZ). It was however demonstrated that the approach is valid with a newest generation DLE industrial gas turbine burner, where a very high degree of mixing between the fuel and the air is achieved with both decomposed NH_3 blends (NH_3-H_2-N_2) or NH_3-CH_4 mixtures [19,20]. In Ditaranto et al. 2022 it was shown that de-rating severely the thermal intensity (MW/m^3/atm) of the combustor under pressure, 100% NH_3 combustion could be stabilized, with promising NOx emissions below 200 ppm @15vol% O_2 with a primary zone equivalence ratio of 1.3 and a combustor pressure of 5 bar.

In Ditaranto et al. [16] a series of results with a non-premixed aeroderivative type burner engine showed an interesting result with NH_3-CH_4 blends as fuel. Although the pressure effect on NOx was still important in the range 1 to 5 bar at all PZ ER, the benefit of burning in a staged combustor with a rich PZ almost disappeared as the NOx emissions measured at the combustor exit were fairly constant over the PZ equivalence ratio range. This is in not in agreement with the results reported in Okafor et al. [14] where measurements with a non-premixed combustor exhibited a clear drop in emissions when operated with a rich PZ. There are turbulent mixing interactions in practical burners that incite to caution when extrapolating findings of NOx behaviour from purely kinetic studies. Other works cited in Table 1 with generic swirl stabilized burners have been performed under relatively low pressure, and the effect of pressure itself was not analysed in details [18,22,23]. To the knowledge of the authors, no detailed studies of application-relevant flames at pressure exceeding 6 bar (cf. Table 1) have been reported apart from the few configurations in the author's own work. There is therefore a critical need of documented experimental studies against which theoretical and numerical studies can compare to.

Duynslaegher et al. [25] conducted pioneering numerical studies that showed the favourable impact of pressure on NO formation. Despite being based on a kinetic mechanism not specifically designed for NH_3, the study revealed a distinct trend of reduced NO levels with increasing pressure. This suggests that it is the central intermediate reactions involved in NH_3 oxidation that exhibit a sensitivity to pressure. The strong effect of pressure is through the control of the

concentration of key radical species affecting the oxidation steps of NH_i intermediates [26–28], and particularly the three-body reaction $OH + H + M \rightarrow H_2O + M$ which increasingly consumes H and OH with pressure, and in turns limits the formation of HNO through NH and NH_2, hence the major intermediates responsible for NO. Although pressure has the opposite effect with hydrogen, for decomposed ammonia blends containing both fuels, the trade-off is in favour of decreasing NOx formation because NH_3 generates NO at two orders of magnitude higher than does H_2. This phenomenon was clearly observed and quantified at moderate pressure increase up to 4 bar in our previous works [16,19,20] with decomposed NH_3 and NH_3-CH_4 blends and both premixed and non-premixed burner configurations. However, the NOx emissions sensitivity to pressure seemed to decrease when approaching 4 bar. In Pugh et al. [17] the pressure was varied up to 6 bar with a non-premixed swirled burner in overall lean combustor condition. Unlike the above mentioned studies of Ditaranto et al. they observed that the NOx reducing effect of increasing the pressure depended on the NH_3 content of the fuel. However, it is suspected by the authors themselves that the combustion efficiency was not complete. They also showed that the flame structure, controlled by 2 different swirl numbers, had a strong impact on both the NOx performance and its dependency to pressure. A gap in knowledge therefore exists regarding the sensitivity of NOx emissions to pressure change beyond 4 bar. This study aims to fill this gap by providing experimental results of ammonia fuel blends combustion from application-relevant type of flames under elevated pressure and diverse combustor operating conditions. The findings also offer valuable insights for gas turbine designers working on combustion systems for ammonia-containing fuels, both in new developments and retrofit applications.

2. Experimental procedure

The burner used in this study and illustrated in Fig. 2 is a SINTEF developed swirl stabilized burner which presents generic features of rapid mixing with swirl stabilization, common in modern gas turbine burners. A central tube supplies the gaseous fuel at its end through multiple radial injection holes in the surrounding air flow. The primary combustion air is supplied around the fuel tube as an annular flow and passes through an axial swirl module with 40° angle vanes, characterized by a Swirl Number of 0.74 based on Syred and Beer [29] geometrical calculation. The annular air channel ends with an outer convergent exit section. The fuel being injected close to the burner exit plane provides some degree of partial premixing. The SINTEF burner has been characterized previously with methane as fuel in oxy-fuel oxidizers [30]. Due to the fuel injection arrangement and swirl configuration, the burner can be characterized as rapid mixing and not fully premixed as although the injection velocity is high the fuel jets
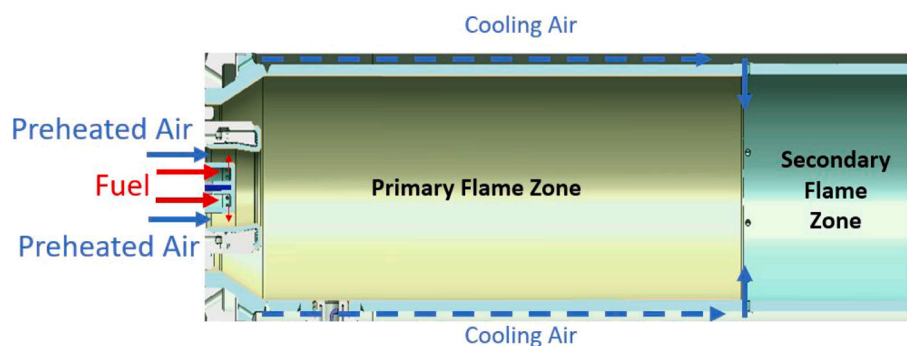
Fig. 2. Burner and combustor arrangement of the HIPROX facility.

do not have time to fully mix with the air before the axial recirculation zone generated by the swirl to stabilize the flame.

The burner was placed inside the high pressure combustion facility HIPROX (part of the ECCSEL European Infrastructure network). The facility consists of a pressure vessel containing the combustor depicted in Fig. 2 and a gas flow and temperature control unit. For this study based on emissions characterization, the combustor was a metal tube with Thermal Barrier Coating (TBC), close to real gas turbine application in terms of wall temperature and heat transfer properties which are known to affect overall emission performance. The combustor has a double wall construction with a flow of air in between for convective cooling purposes. The end of the primary flame zone (PZ) is delimited by the location at which part of the cooling air is injected and mixes with the flame gas products, marked by arrows in Fig. 2. Based on the local momentum ratio in hot conditions, these multiple secondary air jets have enough momentum to penetrate through the whole radius of the combustor, ensuring good mixing with the PZ flame products. The secondary zone (SZ) acts either as a dilution section when the PZ is operated fuel lean (representing conventional gas turbine flame tube operation), or as a secondary flame zone of a staged combustion strategy when the PZ is operated in fuel rich conditions. At the end of the SZ, the rest of the cooling air is injected into the exhaust gas, prior to a water-cooled back-pressure valve that controls the pressure in the combustor. The cooling air is controlled and injected into the plenum, formed between the flame tube and the outer pressure vessel, before entering in the double wall space around the flame tube. Because the burner of this study is not of a fully premixed one, the given equivalence ratio (ER) of the different flame zones refers to the global fuel - air conditions in the zone and is an indication of the availability of oxygen. It is understood that there is a wider distribution of local ER throughout the flame, biased towards richer conditions than the ER indicates.

The global equivalence ratio (PZ + SZ) was always kept within 0.4–0.7. The exhaust gas temperature was measured by a single radiation shielded thermocouple located at the end of the secondary zone. Therefore, the gas temperature measured corresponds to that of the flame products at the global equivalence ratio. The range of experiments related in this study is given in Table 2 and was limited by flame stability, particularly at low partial decomposition (cracking) ratios.

The hydrogen and nitrogen gases composing the fuel are stored in high pressure cylinder racks, while the ammonia is stored as a liquid in a tank before preheating through an evaporator. The air is supplied by the compressed air network boosted to high pressure. The burner air is preheated to temperature specified in the figures, while the cooling air is injected at room temperature. All gas flow rates are controlled by commercial mass flow controllers (MFC) calibrated for the specific used gases before being mixed and supplied to the burner central fuel tube. The mass flow rates uncertainties propagate on the input power and equivalence ratio. Doubling the MFC uncertainties given by the manufacturer (to be conservative), leads to a maximum uncertainties

of 3% and 8% respectively. The corresponding uncertainty on the decomposition ratio (DR) increases from high to low DR with less than 4% above 0.5 and up to 10% at 0.15 (lowest DR achievable above blow off limit).

The exhaust gas was sampled downstream of the back-pressure valve and drawn into a FTIR analyzer (model Gasmet DX 4000). Several species are analysed, amongst others: NO, NO_2, N_2O, NH_3, H_2O. The instrument also provides O_2 concentration measured with a Zirconia cell. Even though the instrument allows to measure in the wet sampled gas, most of the measurements presented have been made in dried sampled gas in order to improve the accuracy on the NO measurement which spectral region of interest overlaps with the strong absorption bands of water. At regular time intervals the sampling was switched from dry to wet to check the measured water content against the expected theoretical mass balance. Owing to a longer residence time in the dry sampling line, the NO to NO_2 conversion rate is expected to be less than 2% and up to 14% for NO concentrations lower than 400 ppm and at 3000 ppm respectively. However, calibration tests with synthetic mixtures of NO, NO_2 and steam relevant to the emissions in this study (up to 4000 ppm), showed much lower conversion rates, if any at all. Practically, it is advisable to exercise caution when using FTIR gas analyzers with gases containing NOx concentrations beyond the calibration range. While weak extrapolation errors may occur for NO, the presence of strong non-linearities makes measurements for NO_2 very erroneous. For the measurements presented in this study, spectra for NO and NO_2 up to 4000 ppm concentrations were prepared and added to the FTIR spectral library, covering thus the entire range. For N_2O calibration spectra in a limited range 50 ppm were only available, higher measured values are commented with the necessary precautions. The uncertainty on the species concentration measurement itself is given by the instrument manufacturer to be within 2%, however uncertainties generated by using an FTIR setup for conventional combustion without the care described above would lead to much higher uncertainties.

All emissions are given corrected to the reference O_2 concentration of 15% and dry basis, as commonly used for gas turbine emissions. Since varying the decomposition ratio of ammonia affects the fuel LHV, this conventional reference correction does no longer allow for a fair comparison. NOx should then be better expressed in mg/MJ, as sometimes found for emission limits in regulations. Owing for the change in fuel LHV, flames with the same NOx emission value corrected to the 15% O_2 dry reference would emit linearly less NOx expressed in mg/MJ as the fuel is decomposed. Up to a 12.7% decrease between pure ammonia and its fully decomposed blend of hydrogen and nitrogen. Because this study focuses on the formation of NOx from flames and the influence of the fuel composition, the use of corrected volume fraction is a more convenient way to express and compare the rates of formation and destruction. Nevertheless this dilution effect is discussed when needed.

Numerical kinetic calculations in a Continuously Stirred Tank (CST) reactor or premixed laminar flow have been performed to support

Table 2
Experimental scope of the study.

Pressure	NH₃:H₂:N₂	Power	Air preheat temperature	Equivalence ratio
1–10 bar	(1-x):(3/2x):(1/2x) $x \in [0, 1]$	10–100 kW	260–310 °C	PZ: 0.6–1.7 Glob.: 0.4–0.7
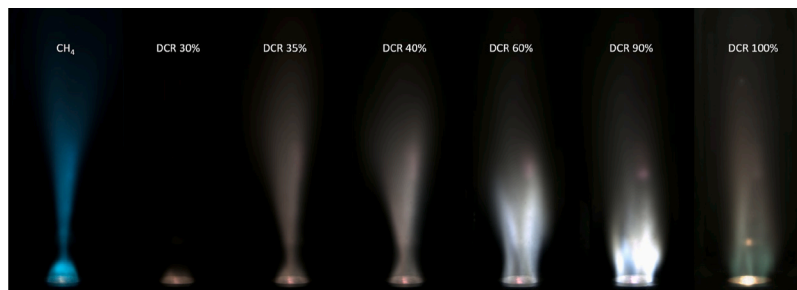
Fig. 3. Direct photographs of flames at 10 kW, ER = 0.8, and atmospheric pressure with various fuels: methane (left) and different partial decomposition ratios of ammonia up to fully decomposed (30% case is an unstable flame).

the analysis of the results by using the CANTERA code [31] with the chemical mechanism of Stagni et al. [32], shown to perform well with NH₃ containing fuels [7]. These simulations do not aim at simulating the flames of the present study which are not fully premixed, but only to isolate the effects of kinetic in the phenomena observed.

3. Results and discussion

Fuels containing ammonia and hydrogen exhibit flames with a broad range of radiative emission characteristics, affecting their visual appearance compared to conventional hydrocarbon flames. In Fig. 3, direct photographs display sample flames investigated at varying decomposition ratios (DR), alongside a reference methane flame using the same burner. It is important to note that these flames were captured in an optical combustor of larger diameter than the metal combustor used for all emission results in this study (cf. Section 2 for details). Additionally, the colour representation in these images is highly dependent on camera settings, which were not identical for all pictures, potentially leading to misleading interpretations.

The primary purpose of these photographs is to provide an impression of the flame structure resulting from a robust swirling flow and combustion intensity facilitated by the increased content in hydrogen. As the decomposition ratio increases, a noticeable trend emerges: the flames become shorter and intensify closer to the burner exit. This phenomenon is attributed to an overall increased reaction rate due to increased hydrogen content in the fuel, enhancing the flame ability to withstand higher strain rates and velocities. Observations reveal that at low DR, stabilization is controlled by the recirculation zone sitting at the burner tip, progressing into a plume-like region after the neck which marks the location of the stagnation point. Contrarily, at high DR the flames manage to anchor themselves around the recirculation zone, featuring visible partially premixed fuel jets. At the lowest DR shown in Fig. 3, the flame is unstable and on the verge of blowing off.

3.1. Single stage combustion mode

3.1.1. Effect of pressure on highly decomposed ammonia blends

At full decomposition of ammonia (DR = 100%), the fuel blend is composed of H₂ and N₂ in 75:25 volumetric proportion. Hydrogen is known to be a fuel with a propensity to form NOx first of all due to the effect of its high adiabatic flame temperature on the thermal NO mechanism. Increased pressure has a tendency to further increase NOx for most fuels through an increased weight of the N₂O pathway. Indeed, formation of N₂O is pressure dependent with the three-body reaction

N₂+O(+M) = N₂O(+M), and further combines easily with other radicals to produce NO and NH (also producing NO). The NNH pathway controlled by O and H is also important and links to the same three-body reaction involving N₂O. Recently, another route has been seen to be important at high pressure through the HNNO intermediate [33]. For hydrogen flames the pressure sensitivity is higher than for most other fuels, one reason is purely kinetic through the higher availability of H atoms in the aforementioned mechanisms, but can also be due to a Lewis number induced thermodiffusive instabilities observed in DNS results [34,35] generating strongly corrugated flames and responsible for local super-adiabaticity. The measurement in the 2 to 10 bar range with the decomposed ammonia fuel shown in Fig. 4 for two equivalence ratios confirm this trend for the lean swirled flame generated by the burner of this study. The marked increase on NOx roughly doubles from 2 to 10 bar and follows reasonably well a power law with an exponent value of approximately 0.5 in the range investigated, in agreement with reported behaviour [36]. The effect of 25% nitrogen dilution combined with the rapid mixing nature of the burner have a beneficial impact on the thermal NO route, explaining the rather good NOx emission performance. In comparison, optimized burners in industrial conditions have shown emission levels of 100 ppm at 15 bar [37]. The effect of adiabatic flame temperature on the thermal NO route is well observed with the clear increase in NOx when the equivalence ratio increases from 0.61 to 0.71 which is accompanied by an increase of ca. 160 K in flame temperature.

When ammonia is not fully decomposed, NH₃ is present in the fuel blend and affects the NOx formation dramatically even at low concentration. At 90% DR (i.e. 5.3% NH₃ in volume in the fuel) and atmospheric pressure, NOx rises in excess of 3000 ppm. Pressure also has a strong effect on NOx formation with ammonia as clearly observed in Fig. 5. Although the Reynolds number varies from 25,980 to 110,610 at 2 to 9 bar respectively, turbulence is high and flame aerodynamic and the flow recirculation structure generated are assumed to be similar because the burner exit velocities and residence times in the combustor are almost constant. The adiabatic flame temperature is pressure dependent, but varies moderately with a 33 K difference between 2 and 9 bar. Therefore, it can be assumed that the behaviour observed in Fig. 5 shows the sole effect of pressure on the NOx formation. This effect is strong from 1 to 5 bar with almost a 4-fold drop in NOx emissions, despite an increasing flame temperature. Indeed, due to the increased thermal energy density, the radiative heat losses are not constant in the experimental series shown in Fig. 5. The measured exhaust gas temperature at the end of the secondary stage increases when increasing pressure, indicating a decrease in heat losses in percentage of input thermal power (temperature difference of approximately 200 °C
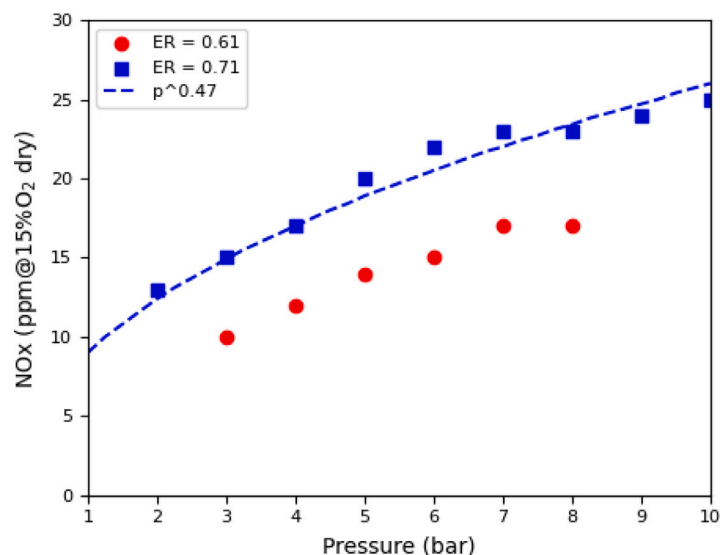
Fig. 4. Effect of Pressure at constant PZ residence time on NOx emissions for 100% decomposed ammonia fuel, air pre-heat temperature 290 +/−20 °C.

between the 2 and 9 bar cases). Therefore, combining a lower heat loss and an increase in adiabatic flame temperature, the flame is hotter with increasing pressure, yet NOx decreases through a stronger kinetic effect. The positive trend in NOx emissions with pressure does however not persist beyond 5 bar and remains almost insensitive to a further pressure increase, at least up to the 10 bar limit of our study. These results confirm the trend observed in the same experimental facility with industrial gas turbine burners (DLE and non-premixed), but limited to 5 bar and with various ammonia fuel blends [16,19,20]. For comparison, Fig. 5 also shows the measurements from Pugh et al. [17] obtained from a non-premixed burner with an overall ER of 0.55 up to 6 bar. Even though the NH_3 content of the fuel is higher, NOx values are significantly lower than this study, however it is suspected by the authors that the combustion efficiency was not complete and that there was NH_3 slip (not measured). This is indeed a general behaviour observed in most studies showing relatively low NOx levels: they are generally accompanied by either or both NH_3 slip and high N_2O emissions, making the results little relevant as far as the application is concerned. It is seen in Fig. 5 that a power law with a negative exponent empirically describes the general behaviour. The scaling factor (3077 in this case) is burner and operating conditions dependent while the exponent, which determines the plateau level, is not thought to be constant as it will be explained later.

The lines in Fig. 5 are results from calculations in a CST reactor with 20, 40, and 80 ms residence times. The kinetic mechanism used [32] reproduces well the pressure effect which is steeper in the first few bars and flattens thereafter, but not the magnitude of the reduction. These calculations are not meant to represent the conditions in the present flames, in fact they are used to isolate the chemistry and show that phenomenon observed is dominated by kinetics, therefore the absolute values cannot be compared to the experimental ones. It will be shown in the sections to come that this transition pressure is not an absolute value and depends on other factors.

This effect of pressure has therefore the opposite behaviour of that seen for the fully decomposed ammonia, despite hydrogen having the largest concentration in the fuel (71% in the 90% DR case). The 5.3% volume of ammonia in the fuel is responsible for the largest quantity of NOx through fuel NOx, and largely outweighs the increasing production of NO through the thermal route linked to the presence of H_2. The NO_2/NO ratio shown in Fig. 5 also decreases with increased pressure following the same trend as NOx, although the NO to NO_2 conversion is generally favoured by pressure in presence of oxygen. Therefore, this indicates that the phenomenon that leads to increased NO_2 fraction of NOx at lower pressure is driven by the NO mechanism itself in the flame zone and not a post-flame oxidation.

3.1.2. Effect of decomposition ratio

The previous section highlighted the strong and dominant conversion rate of NH_3 to NOx in lean conditions and the positive effect of pressure in the case of low NH_3 content in the fuel (high DR). Fig. 6 shows now how increasing NH_3 in the fuel – through a lower decomposition ratio – affects NOx formation. For all the data points shown in Fig. 6, no NH_3 slip were observed in the exhaust gas, indicating that these operating flame conditions were stable, except when approaching for the "blow off" points marked accordingly in the figure. At both 1 and 5 bar pressure the stability limit at low DR is at approximately 20%, corresponding to a minimum of 25% H_2 in the fuel blend. The pressure dependency is again well marked with NOx concentration peaking at nearly 6000 ppm at 50% DR at 1 bar against 3200 ppm at 5 bar (a halving) at the same equivalence ratio. The strongest relative effect is however not at 50% and increases with DR with a 4-fold reduction of NOx at 90% between 1 to 5 bar.

The second observation is that the NOx behaviour is not monotonic with the fuel NH_3 content, but peaks at mid DR value of 50%, corresponding to a fuel mixture 33% NH_3, 50% H_2, and 17% N_2 by volume. Wang et al. [18] observed a similar phenomenon with a NH_3-CH_4 fuel blends, but peaking at 30%–40% NH_3. When the initial concentration of NH_3 in the fuel increases, it is expected that NOx increases, as it is the case in the high DR region of Fig. 6. Note that at full decomposition (DR = 1) NOx values are below 30 ppm although they appear as zero due to the large scale of the plot. When DR decreases, the adiabatic flame temperature decreases due to the lower content of hydrogen in the fuel mixture, thus moderating the increasing NOx formation due to the said increasing NH_3 content. Indeed, there is approximately 150 °C difference in adiabatic temperature between 100% and 20% DR. To show the extend of that trade off, the results of two kinetic calculations in a CST reactor are plotted in Fig. 7, corresponding to the same inlet conditions as in the experiments of Fig. 6. For each pressure, two cases are presented: one corresponding exactly to the ER = 0.8 and pre-heated air temperature conditions of Fig. 6, and for the other (in dashed line) the inlet air temperature is adapted to give a constant adiabatic flame temperature throughout the DR range. The reactor residence time is longer than in the PZ of the experiments of Fig. 6 and has been selected to be long enough to give full consumption of NH_3 as in the experiments (note however that the trend and conclusions drawn were not affected by selecting a shorter residence time in the CST reactor, it just lead to NH_3 slip in the calculations). It is observed that when the adiabatic temperature is kept constant (dashed lines), NOx do indeed increase monotonically as NH_3 in the fuel increases (i.e. as
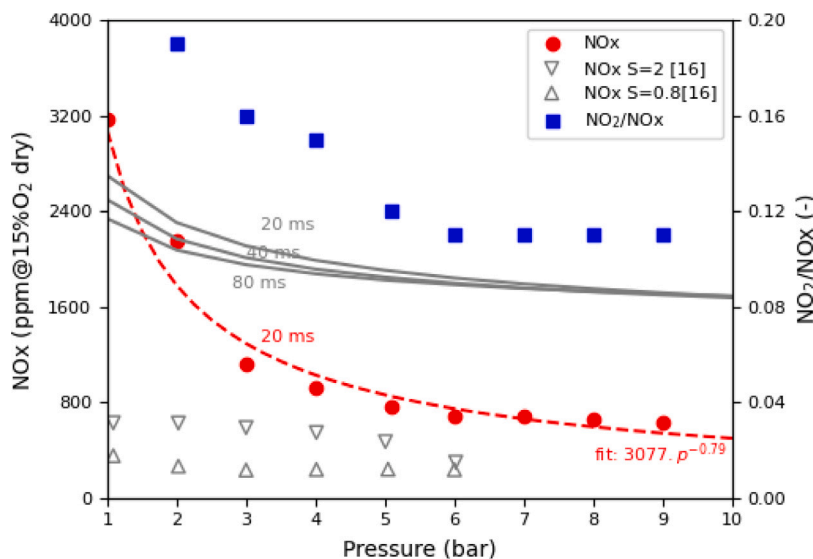
Fig. 5. Effect of Pressure at constant PZ residence time on NOx emissions for 90% decomposed ammonia fuel, ER = 0.7, and air pre-heat temperature 290 +/−20 °C. CST reactor simulations (grey lines) at same input conditions, but varying residence times. Data from Pugh et al. [17] in non-premixed conditions with $0.25NH_3/0.75H_2$ fuel and ER = 0.55.
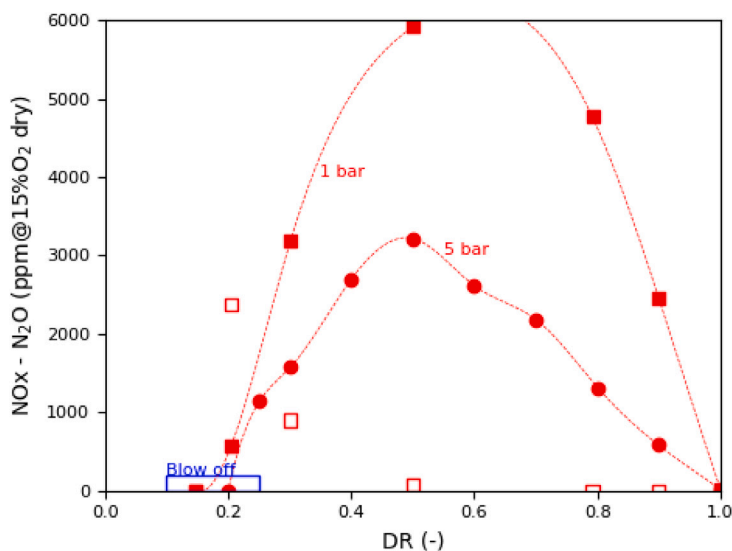

Fig. 6. Effect of DR on NOx (filled symbols) and N_2O (empty symbols), and blow off limit at ER = 0.8. Combustor pressure/air pre-heat temperature: 1 bar/210 °C (squares) and 5 bar/290 °C (circles). N_2O at 5 bar are below 1 ppm and not shown. Data points $N_2O \gg 50$ ppm have higher uncertainty. No NH_3 slip were detected in any of these experiments.

DR decreases). There is therefore a kinetic trade-off between the flame temperature and the NH_3 content in the fuel which is, at least partly, responsible for the non-monotonic dependency of NOx formation to the NH_3 content in the fuel observed both in the experiments and reproduced by the calculations. The mismatch in the peak position between experimental and numerical results can be due to the kinetic mechanism, which is still an active area of research particularly in high pressure conditions, but also due to the complex turbulent flame structure which deviate from a simplified CST reactor. Indeed, the calculations do not show flame extinction at low DR, nor was is the aim of the exercise, because these types of reactor only consider kinetic without any effects of strain rates and mixing.

Mei et al. [3] also found a non-monotonic behaviour of NOx in the post flame region of a 1D laminar flame simulation against decomposition ratio, with a NOx peak value located at around 70% decomposition. They claim that it is the result of a trade off between the increase of O and OH radicals when H_2 increases (which react with NH*i* to form higher NO), and the drop of NH_3 mole fraction that results in rapid decay of its decomposition products, leading to the reduced NO

formation. In fact, their calculations used a fictive species method to decouple the thermal effect and found that, unlike in our calculations, the thermal effect was minor and only localized at the peak region. We argue that the reason for this discrepancy between the studies lies in the time scale of the 1D laminar flame which has a much shorter residence time in the high temperature zone than in our calculations which have residence times relevant to practical gas turbine combustors. Shorter residence times limit greatly NO reduction in NH_3 rich combustion, as it will also be discussed later. In fact, the peak NO concentrations calculated in Mei et al. were in the range of 14 000 ppm, which is one order of magnitude higher than both calculated and measured in our experiments in a flame configuration.

The decomposition ratio impacts also the formation of the potent greenhouse gas N_2O with a sharp increase as DR decreases. The N_2O concentrations in Fig. 6 were actually measured, but the calibration spectra in the FTIR analysis were limited to a lower range, therefore there is a high uncertainty on the actual values, but there is no doubt about the sharply increasing concentration trend as the IR signature of N_2O was clearly present in the spectra and its absorption strength
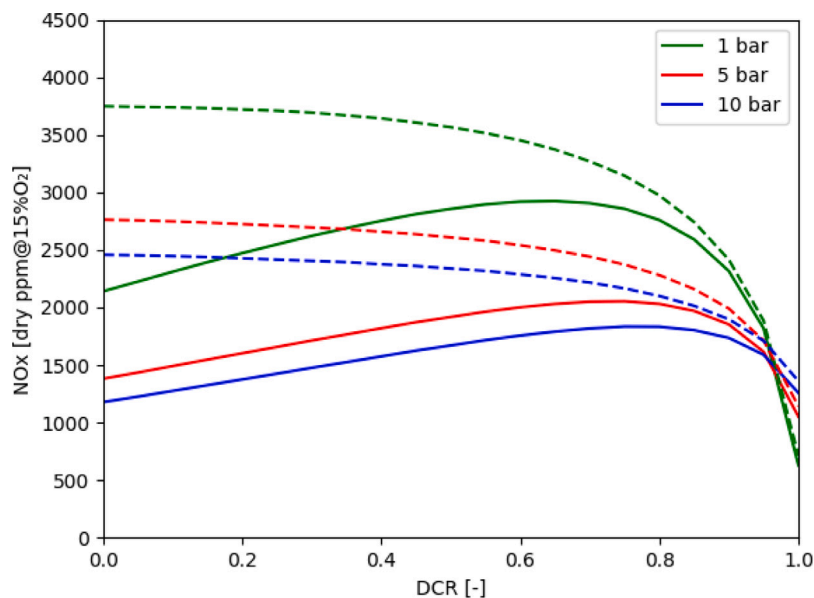
Fig. 7. Calculated NOx vs DR in CST reactor with inlet conditions equivalent to Fig. 6 (lines) and by keeping constant reactor adiabatic temperature (dashed lines). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

greatly increases. As observed for NOx, pressure has also a positive effect in limiting the formation of N_2O, with only a moderate increase reaching 10 ppm in the 5 bar case, just before the DR case that blows off is reached. This behaviour of high N_2O when approaching lean stability limit seems to be reproduced in many studies [15,19]. Kinetically, N_2O formation is little pressure dependent [38], therefore it is believed that the reduction of its concentration with pressure is governed by better stability and therefore combustion efficiency performance, induced by increased turbulent flame speed with pressure. This correlation between N_2O and stability of turbulent flame will be further discussed in the next sections.

3.1.3. Effect of residence time

It is important to understand the conditions that make the kinetics of NH_3 combustion to fully proceed to ideally nitrogen and water. The residence time in the primary zone, an important parameter in combustor system design, has been varied in the experiments leading to the results of Fig. 8 by increasing the flow rates at the burner, while keeping a constant pressure of 5 bar and a constant equivalence ratio of 0.8. To provide a reference, conventional gas turbine combustor have typical primary zone residence time in the range of 20 ms. Two DR cases have been chosen, one on each side of the NOx peak of Fig. 6 and are seen to behave in opposite trends with regards to NOx formation. At high DR the increase in residence time is beneficial in favouring the reduction of NO, as a result of kinetics alone when NOx is dominated by fuel-NO production. This is expected because in the kinetic route for oxidizing NH_3, NO is a natural and inevitable intermediate, as shown in Fig. 9 where after the formation of NH_2, the largest fluxes go through NO. Therefore an increase in residence time in the high temperature region pushes the reactions towards N_2, and this effect is clearly captured in the CST calculations plotted in Fig. 8 as lines. Due to the scale of NOx produced, the NO formed through the Thermal kinetic route (which on the contrary increases with increased residence time in the hot flame zone) may alter, but does not dominate this behaviour.

The trend is however different for the 25% DR case, and at first surprising since it contains even more NH_3 in the fuel, and indeed the behaviour is not reproduced by the calculations. The difference is attributed to a structural effect overtaking the kinetic behaviour, which is a common phenomenon in applied flames where non-linear chemistry - turbulence interactions exist. With a high H_2 content, the 90% DR flames are not subject to stability issue even at high burner

exit velocities (i.e. low residence times) and are therefore rather kinetic controlled. But for the 25% DR case, the flame speed is far slower (calculated laminar flame speed is 21.9 cm/s and 210.8 cm/s for 25% and 90% DR respectively) and affects the flame structure strongly, as seen in the flame photographs (cf. Fig. 3), where at high DR the flame stabilizes much closer to the burner. The flame stability is, as a consequence, impaired inasmuch as blow off is eventually reached if the burner velocity further increases (see mention in Fig. 8). At these high burner velocities and high strain rates in the swirling flow, it is expected that NH_3 slips through the PZ, and it is suspected that it participates to de-NOx reactions in the SZ which is a region with proper temperature window. Consequently responsible for the combination of both lower NOx and absence of NH_3 in the exhaust gas. This is a reasonable assumption as Osipova et al. [39] measured non-decomposed NH_3 surviving through an equimolar NH_3-H_2 laminar flat flame front zone of ca. 1800 K. In these conditions, NOx increase with residence times, or rather with lower burner exit velocities, because of better flame anchoring and NH_3 combustion efficiency. Nevertheless it is expected that by further increasing the residence time in these experiments, NOx would have started to decrease, as in the 90% DR case. In fact, Wang et al. [18], with a 80% NH_3 fuel content observed a reduction in NO by increasing the residence time through both an increase in swirl number or chamber length, as kinetically anticipated, but their burner exit velocity was 5.4 m/s, which is still higher than the slowest exit velocity of the results reported in Fig. 8, therefore with lower turbulence and strain rates, hence better flame stability.

The other observation of interest from Fig. 8 is that for the 25% DR case, N_2O increases in concentration as soon as stability issues arise. This is in line with the above analysis, as in the NH_3 oxidation chemistry, N_2O forms and peaks early in the flame front [39,40], before NO starts building up. Therefore it is not surprising that N_2O is formed rapidly at the early steps of oxidation and then quenched by straining in the flame, which at the same time prevent full NH_3 oxidation from proceeding. Note that due to the high greenhouse gas potential of N_2O, it only takes 200–300 ppm in typical gas turbine exhaust gas to have the same global warming effect as if natural gas was used. The results confirm that NH_3 requires long residence times at high temperature to maximize reduction of NOx and N_2O. This was also clearly evidenced in Ditaranto et al. [19] where by de-rating the combustor design thermal intensity, not only a 100% NH_3 flame became stable thanks to the associated low burner velocity, but around 200 ppm @15vol% O_2
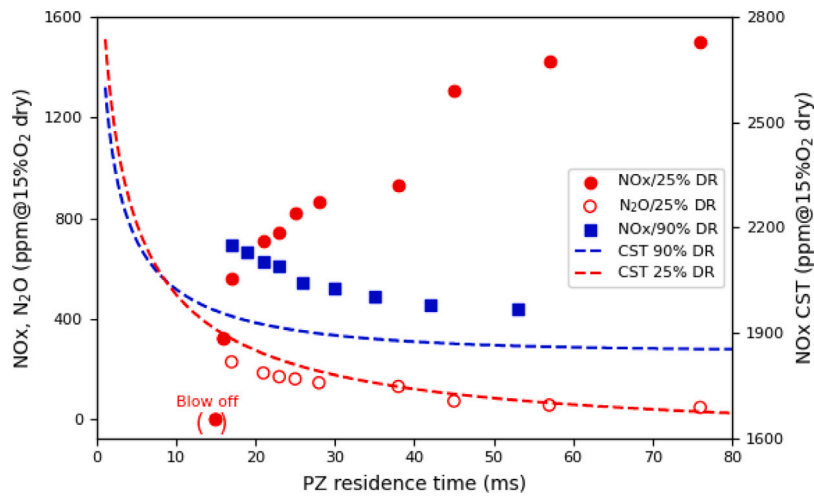
Fig. 8. Effect of PZ residence time on NOx, N_2O, and blow off limit at constant pressure (5 bar), air pre-heat temperature (280 +/−25 °C), and equivalence ratio (0.8). Symbols: 25%DR (circles) and 90%DR (squares); NOx (filled) and N_2O (empty). Data points $N_2O \gg 50$ ppm have higher uncertainty. Dashed lines: CST calculations. At 90% DR $N_2O = 0$ and not shown.
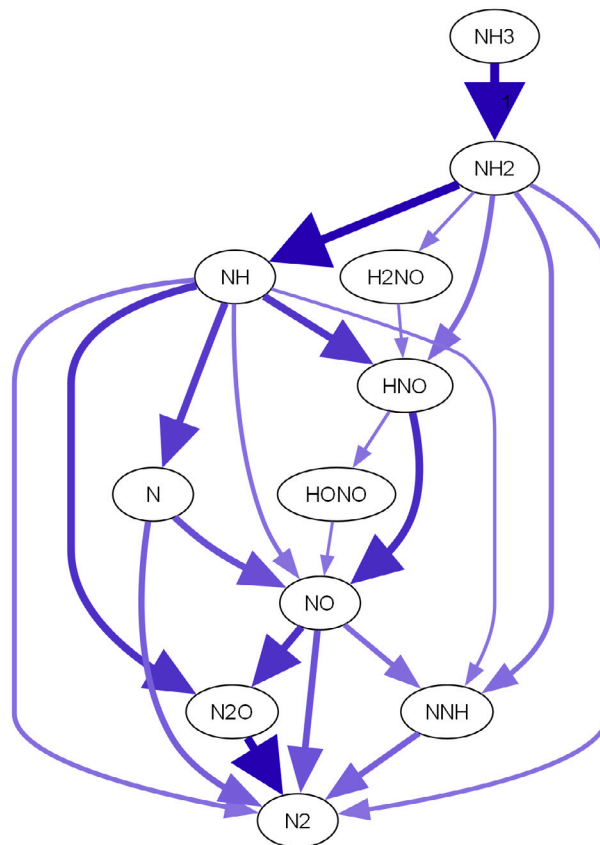

Fig. 9. Kinetic path following N-atom for a 25% DR fuel blend at 5 bar, equivalence ratio 0.8, and air pre-heat temperature of 280 °C.

with a PZ equivalence ratio of 1.3 and a combustor pressure of 5 bar was achieved. We therefore conclude from Figs. 6 and 8 that most turbulent flame conditions leading to a NOx decreasing behaviour when increasing either or both the NH_3 fuel content and the burner exit velocity, is often associated with a loss in flame stability and increase in N_2O, and cannot therefore be used as a primary NOx abatement strategy in combustor design. These experimental observations stress the importance of considering the turbulent and dynamic conditions at burner exit when dealing with practical flames and not only kinetic considerations, especially with low reacting fuels like ammonia.

3.2. Two stage combustion mode

3.2.1. Effect of primary zone equivalence ratio in two stage combustion

To minimize the formation of NOx with ammonia as fuel, staging the combustion by keeping the primary zone fuel rich can be a promising strategy [27]. Fig. 10 showing emissions measured at the end of the secondary zone, is a clear evidence of that effect at both low (25%) and high (90%) ratios of decomposition. Lower O_2 availability limits the oxidation steps and NH_3 preferably decomposes into stable H_2 instead of forming NH*i* radicals that are responsible for further leading
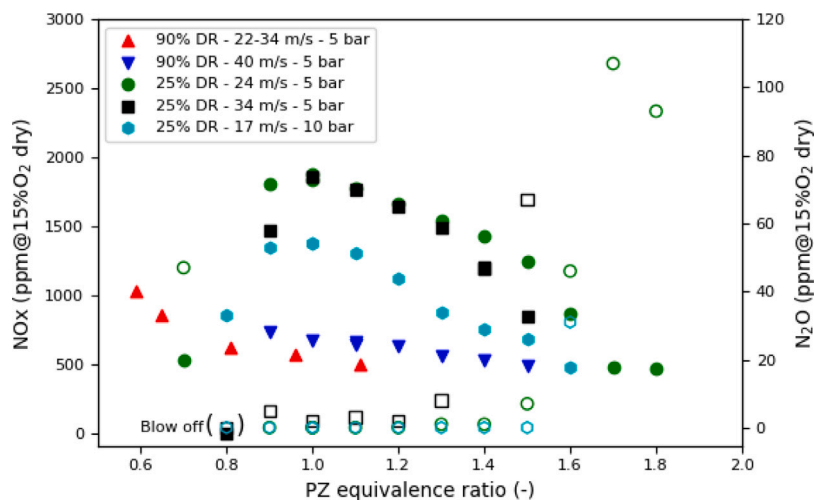
Fig. 10. Effect of PZ equivalence ratio on NOx and lean blow off limit at 90% DR and 25% DR. NOx: filled symbols; N_2O: empty symbols. Data points $N_2O \gg 50$ ppm have higher uncertainty.

to NO. Then, in the secondary zone a higher share of the production rate of NO becomes controlled by temperature (thermal NO route) at a much lower rate. Many studies have demonstrated that effect, but the following results stress again the importance of considering flame — mixing interactions when dealing with turbulent flames. Consider the results for the 25% DR case at 5 bar at two burner velocities shown in Fig. 10, where the flames at the highest velocity are at the limit of stability and often trigger a thermo-acoustic instability. The NOx emissions in both cases are only matching close to the stochiometric region, but decreases quicker on both lean and rich sides for the high burner velocity (most unstable) case. This decrease, accompanied by a rise in N_2O, is attributed to a loss of combustion efficiency as the flame speed properties are getting poorer, therefore less fuel NH_3 is converted to NO and is escaping unburned into the SZ and either participating in NO reduction or slipping out. This behaviour is supported by CST calculations showing that approaching lean conditions, full consumption of NH_3 becomes difficult. Comparing 5 and 10 bar results shows that the positive impact of pressure on NOx discussed in the previous section in lean conditions holds when the PZ is rich.

Considering now the 90% DR case, when the equivalence ratio is varied at both constant velocity or constant power, there is no lean side reduction of NOx as observed at low DR, which is attributed to a better flame stability and higher temperature at high H_2 content fuel. In these conditions, NH_3 is fully consumed and the leaner the PZ, the higher the NOx as kinetically expected. The NOx reduction by operating with a rich PZ is less pronounced, but general lower values than at lower DR are achieved.

3.2.2. Effect of pressure on decomposed blends in two stage combustion

The beneficial effect of pressure of on single stage (lean) combustion is also reproduced in a similar manner in the two stage configuration with rich PZ as shown in Fig. 11, but the effect is highest for low and middle DR values, and not significant when the fuel NH_3 content is low (at 90%DR). In other words the strongest effect of pressure is on high emission values and the effect decreases when NOx concentrations reaches the 500 ppm range. For comparison, the plotted data of Ditaranto et al. [20] obtained with a fuel corresponding to a 22% DR also experience the steep decrease in NOx with pressure. The significant difference in concentration levels with those obtained with this burner highlights the strong burner and flame structure dependency. The fitted power law in the graph is only shown to express the strong, but decreasing rate of the NOx dependency to pressure, and the negative exponent obtained is not universal. To decouple any effect of residence time from that of pressure, two sets of experiments are shown: one

by only varying pressure, all flows kept unchanged, and the other by adapting the flow rates in combination with pressure to keep the residence time (hence burner exit bulk velocity) constant. The latter is of practical significance when gas turbines are changing load (ramping up and down), and was chosen here to correspond to the same thermal intensity ($MW/m^3/bar$) for all three fuels. It can be seen that both increasing pressure and residence time participate in lowering NOx. At 90% DR the advantage gained by operating with a rich PZ does not combine positively with pressure because the NOx concentrations are already in the lower range at atmospheric conditions. For this highly decomposed ammonia fuel, a very large reduction in NOx is achieved by operating the PZ rich already at atmospheric pressure (compare with the results of Fig. 5 for 90% DR with lean PZ).

The beneficial pressure effect on NOx formation is seen to act differently as that observed in Fig. 5 for 90% DR. Whereas for the lean 90% DR case the pressure effect weakens and disappear above 5 - 6 bar, this transition is moved to 8 - 9 bar with the 25% and 50% decomposed fuels. This observation is an indication that it is not a transition linked to an absolute pressure value, but a combination of several factors. In conclusion, it appears that longer residence times, increased pressure, and rich PZ operation are all parameters limiting NOx emissions from all decomposed blends of ammonia, but that they only positively add up to a certain point where a sort of a equilibrium barrier is reached. This NOx plateau level being higher at higher NH_3 content in the fuel.

3.2.3. Effect of decomposition ratio in two stage combustion

As observed in the lean case, the results of Fig. 12 in staged combustion mode with a rich PZ, the NOx performance is not monotonically evolving with the content of NH_3 in the fuel. At the same combustor pressure the NOx levels are lower, particularly for the worst cases at mid decomposition ranges 30%–80%. At higher pressure (10 bar), the NOx peak zone gets lower and flatter for a wider range of DR, which again shows that there is a limit to which it is possible to combine a rich PZ and high pressure to further decrease NOx emissions. The location of the peak NOx is not clear and is believed to be a complex trade off between the flame structure, NH_3 in fuel, and temperature (see discussion in Section 3.1.2) that places the peak NOx at various DR. Another advantage of operation with a rich PZ is that the flame stability is improved and the blow off limit is pushed towards lower DR (down to 10% against ca. 20% when lean). This can be attributed to the laminar flame speed peaking at rich conditions as demonstrated for fuels of various compositions of H_2 and NH_3, e.g. [3,41]. Unlike in single stage lean combustion configuration, there is no tendency to generate N_2O when approaching flame stability limits, as no N_2O
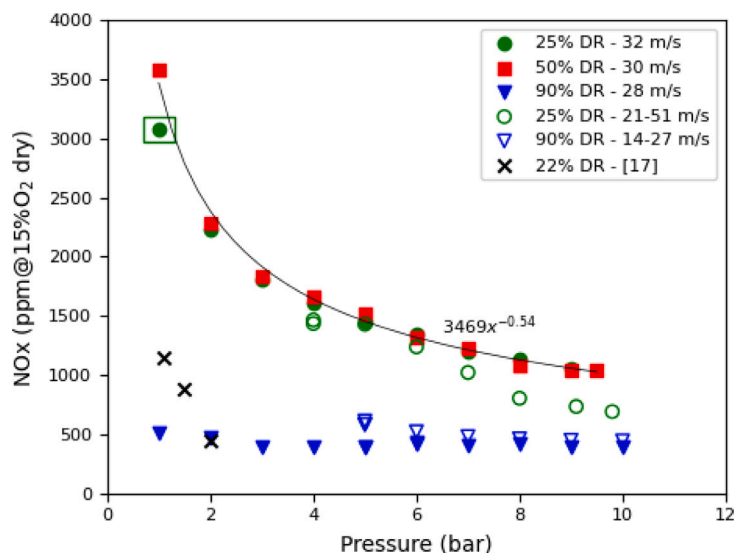
Fig. 11. Effect of pressure at constant and varying PZ residence time on NOx in staged configuration with PZ ER = 1.3 and 290 +/−20 °C air pre-heat temperature. N₂O was only detected on circled point with a value of 213 ppm. Data from Ditaranto et al. [20] obtained with a DLE burner at PZ ER = 1.11.
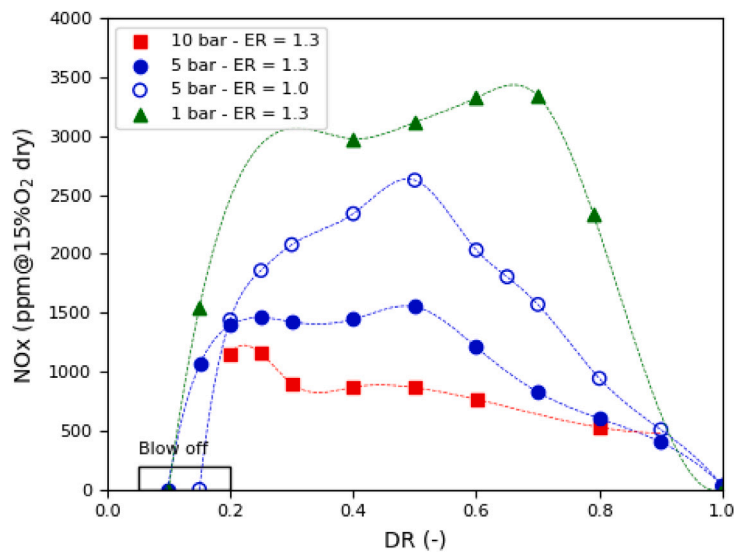

Fig. 12. Effect of DR on NOx and blow off limit. 290 +/−20 °C air pre-heat temperature, except at 1 bar: 210 +/−20 °C. No N₂O detected in any conditions.

was detected in any of the rich PZ operating cases shown in Fig. 12. This is in agreement with the kinetic of NH_3 oxidation and also due to the limited availability of O_2. Note that in Fig. 12 when DR is varied, the fuel thermal power input is adjusted to be kept constant, therefore the NOx emissions in the unit given in the plots would vary strictly proportionally if they were expressed in mg/MJ.

Fig. 13 shows that the relative NOx reduction effect of pressure increases with DR and flattens at around a 4-fold reduction from atmospheric to 5 bar combustor pressure when DR $\geq$ 65 +/- 5% (i.e. lower than ca. 20% NH_3 in the fuel blend). As noted previously this effect diminishes at further pressure increase and for a 10 fold increase in pressure, this ratio is lower at all DR except in the asymptotic plateau which starts earlier, at 55 +/−5%DR (i.e. lower than ca. 30% NH_3 in the fuel blend). Moreover, the plot shows that the pressure effect is relatively the same whether the PZ is lean or rich, which is seemingly in disagreement with measurements made in Osipova et al. [39] in a laminar flat flame conditions. However, the experimental results in Osipova et al. in the range 1 to 6 bar were obtained in the post flame region without secondary combustion zone where they did measure unburned NH_3. With a secondary addition of air, this NH_3 would truly

oxidize to NOx and cancel out the effect and be in better agreement with this measured trend.

4. Conclusion

The study provides experimental findings on the combustion of $NH_3/H_2/N_2$ fuel mixtures, corresponding to varying degrees of ammonia decomposition (cracking). The setup, featuring a swirl-stabilized burner and a combustor operating under high pressure up to 10 bar, simulates conditions relevant to gas turbine engines. The experimental observations indicate that flame stabilization significantly weakens when the decomposition ratio (DR) is below approximately 30% (i.e., ammonia concentration above 70%) for that particular burner. Pressure strongly influences NOx emissions whenever ammonia is present in the fuel blend, experiencing a sharp decrease during the first pressure increase steps, but remaining stable up to at least 10 bar — the limit of this study. The transition pressure at which this effect declines is dependent on other factors like equivalence ratio, DR, and residence time, and is therefore not an absolute value. In the fully decomposed case the trend reverses, as expected for hydrogen fuels. However, NOx
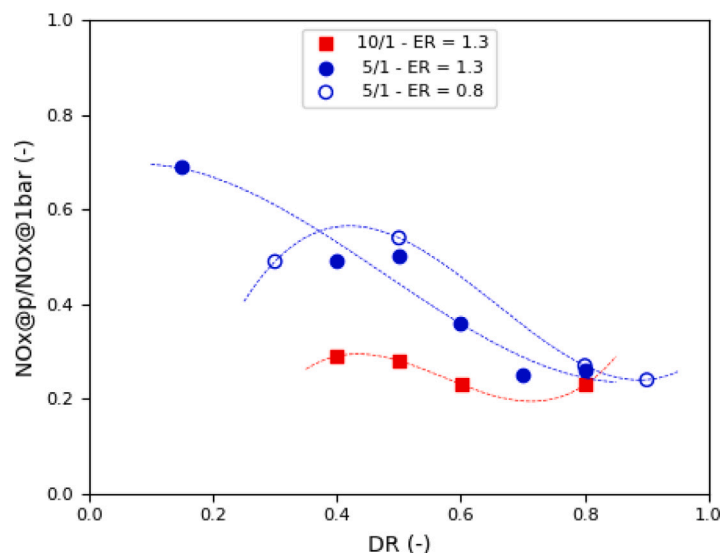
Fig. 13. Ratio of NOx produced at two different pressure at fuels with varying decomposition ratios.

levels in partially decomposed NH_3 fuels are much higher, maintaining a decreasing trend even at high DR close to one.

All else being equal, the propensity for NOx formation is not monotonic with the decomposition ratio, but appears as a trade-off between NH_3 concentration in the fuel and the actual flame temperature. For the burner and combustor configuration of this study, the trade-off peak occurs at around 50% ammonia decomposition (33% NH_3 in the fuel). The pressure increase benefits fuel NOx formation across all equivalence ratios, with the relative decrease in NOx being highest at high DR, stabilizing towards a maximum of a 4-fold reduction at mid and high decomposition ratios.

Operating the primary zone lean for practical turbulent flames shows a decrease in NOx due to a loss of combustion stability and a possible NH_3 slip, reducing NO in the secondary flame zone. Considering the wide parametric variation of this experimental study, it was generally found that all factors tending to weaken flame stability (e.g. increasing fuel NH_3 content, approaching lean/rich blow off limits, or increasing burner exit velocity) favour emissions of N_2O emissions irrespective of PZ ER, whether lean or rich. Therefore, a very lean primary zone is not deemed a viable strategy as a combustor technology, negating the low greenhouse gas benefit of ammonia as fuel. Pressure increase effectively limits this trend, similar to its effect on reducing NOx emissions. As a general trend, it appears that longer residence times, increased pressure, and rich primary zone operation are all parameters limiting NOx formation from all partially decomposed blends of ammonia, but that they only positively add to a certain limit of a few hundreds ppm. This NOx plateau level being higher at higher NH_3 content in the fuel. Additionally, longer residence times moderate the trend, suggesting that decreasing combustor thermal intensity (i.e., $MW/m^3/atm$) could be a way to mitigate in-combustor NOx formation at the expense of larger hardware. Finally, the fuel obtained by perfectly decomposing ammonia appears to be the optimal fuel associating strong flame stability and low NOx propensity, even for the non fully premixed burner type of this study.

CRediT authorship contribution statement

Mario Ditaranto: Designed research, Performed experimental work, Processed and analyzed data, Wrote the paper. **Inge Saanum:** Performed experimental work, Processed data.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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