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Citation for final published version:

Valera Medina, Agustin , Pugh, Daniel , Marsh, P., Bulat, G. and Bowen, Philip 2017. Preliminary study on lean premixed combustion of ammonia-hydrogen for swirling gas turbine combustors. *International Journal of Hydrogen Energy* 42 (38) , pp. 24495-24503. 10.1016/j.ijhydene.2017.08.028

Publishers page: <http://dx.doi.org/10.1016/j.ijhydene.2017.08.028>

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Preliminary Study on Lean Premixed Combustion of Ammonia-Hydrogen for Swirling Gas Turbine Combustors

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Abstract

Hydrogen has been considered one of the most promising materials for energy storage during the last decade with considerable research having been undertaken to demonstrate the use of the molecule in power production systems. However, hydrogen presents drawbacks in terms of global commercialisation and deployment since its distribution is only feasible with significant dedicated infrastructure investment including liquefaction or if it is combined with other gases such as methane. The latter will still produce carbon emissions, whilst the former is not economically viable with current technologies. Therefore, an alternative is to use ammonia as a hydrogen storage vector. Ammonia, a molecule that has been used for more than a century, is a well-known material distributed across the world. Moreover, its properties allow its liquefaction at a relatively low pressure under atmospheric temperature compared to hydrogen, serving as a compound that can be used from fertilising to industrial processes. For power generation, ammonia has demonstrated to have a very slow reaction hence flame speeds, thus one option is to dope the fuel with a more reactive molecule such as hydrogen, which conveniently can be obtained from cracking ammonia. Hence, this paper presents the results of a numerical and experimental campaign where a 50:50 (vol%) ammonia-hydrogen blend was used for lean premixed combustion in a generic swirl combustor used in gas turbine studies. The results show that whilst the mixture can produce a good flame velocity similar to methane with the mixture having near equivalent laminar flame speed characteristics, the high diffusivity of hydrogen under these conditions leads to a narrow operational envelope with the potential for boundary layer flashback. High NO_x emissions are produced due to the excess production of OH and O radicals. Recommendations for further studies and future developments are also discussed.

Keywords: Ammonia combustion, premixing, swirling flows

Introduction

Gas turbines for hydrogen utilisation have been under development over the past decade, with several companies claiming equipment capable of operating with H₂ from Integrated Gasification Combined Cycles (IGCC) [1-2]. However, hydrogen produced from green sources would currently have to be produced near IGCC facilities to avoid mixing H₂ with other HC gases that can potentially produce greenhouse gas emissions. Therefore, whilst hydrogen has the potential to become one of the major fuel players in the near future for power generation, distribution and storage still remain significant challenges. Thus, an alternative way of using hydrogen in these facilities is via hydrogen-containing fuels. Ammonia (NH₃) has different

combustion characteristics compared with conventional hydrocarbon and hydrogen fuels. Ammonia offers a considerable distribution network with knowledge that has developed for over a century or more, thus potentially setting the path towards a “Hydrogen economy via Ammonia”.

One of the main drawbacks of using ammonia for power generation is the production of NO_x due to the nitrogen contained within NH_3 . Moreover, the low flame speed of ammonia leads to early blowoff or difficulty providing ignition, with the consequence of high emissions during the start-up procedure. Considerable research took place to support the development of gas turbine combustion systems during the 1960's [3-5]. These studies demonstrated the low reactivity of ammonia with its ignition energy considerably higher when compared with fossil fuels. Further experiments also showed that the fundamental problem with NH_3 -air blends for gas turbine operation is the relatively slow chemical reaction rate that produces laminar burning velocities of around 6-8 cm/s [6].

However, it was also found that dissociation of the molecule could produce faster flames which could have even similar properties to common hydrocarbons. Results showed that 28% dissociated NH_3 could be used as a substitute fuel in gas turbine combustion systems as the hydrogen enhances reactivity hence increasing burning velocity and reducing ignition energy. Other studies have been conducted to use ammonia as an engine fuel. These studies have succeeded in demonstrating effective power generation using the cracked molecule along with ammonia while falling short in reducing in particular NO_x emissions [7-9]. Other groups, particularly in the USA and Japan, have undertaken studies using ammonia fuelled systems for power production and propulsion systems [10-13], with similar outcomes. Therefore, to develop an efficient gas-turbine combustor which burns NH_3 - H_2 blends whilst maintaining high combustion stability and ultra-low emissions, it is necessary to determine the combustion performance utilising current technologies in order to derive improved designs that can cope with the challenge of using a fast reacting molecule (H_2) alongside a low flame speed composite (NH_3).

Regarding current flow stabilisation systems, swirl stabilised combustion of ammonia with other molecules has been analysed by Meyer et al [14] using swirl stabilisers and a self-sustained heat exchanger for combustion improvement and emissions reduction. They demonstrated that recirculation can reduce considerably the NO_x emissions whilst producing more efficient combustion profiles. A new national program in Japan – SIP (Strategic Innovation Program) - has commenced to decarbonise their energy sector, with support towards the development of new ammonia-based gas turbines. The most recent development is a micro-gas turbine of 50 kW that is run using kerosene-ammonia blends. Although the NO_x levels are still high - up to 600 ppmv - it is clear that the inclusion of catalytic converters can mitigate the problem considerably, thus leading to the production of clean energy if ammonia is recovered via sustainable sources [15-18]. Further studies on pure ammonia combustion using swirling flows [19] or porous media [20-21] have also revealed the potential of ammonia for power purposes at small scale.

Valera-Medina et al. [22] presented studies using a generic swirl burner that was fuelled by a range of ammonia and methane blends. The results showed the subtlety of stabilizing

premixed ammonia blends. It was found that NO_x and CO emissions were considerably low at high equivalence ratios >1.10, identifying a region of chemically reactive balance between methane and ammonia combustion. Xiao et al. [23] have also assessed the potential of using ammonia/methane blends for power generation. Using the Konnov reaction mechanism, a 2D numerical study was undertaken to determine the characteristics of methane/ammonia blends and their combustion characteristics. A reduced mechanism was successfully developed, and utilised to demonstrate good agreement with data, identifying the low NO_x production at high equivalence ratios. Other studies have commenced, developing a new scientific and engineering trend to use ammonia as fuel, enabling the development of “ammonia batteries” to contribute to a decarbonised Europe. Furthermore, multi-national companies such as NUON have also started ambitious programs to develop their capability to operate ammonia-based power plant [24-27]. However, the program has claimed that the limited research into turbine combustion of ammonia has led the consortium to consider cracked hydrogen to fuel their systems.

Thus, this study characterises a 50:50 (vol%) ammonia-hydrogen blend to support the progression towards development of a new generation of gas turbines based on ammonia produced from green hydrogen. The study utilises theoretical calculations to determine regions of low NO_x that could be used potentially for power generation, followed by a 1D numerical analysis to determine the reacting progression of species and an experimental programme that showed the challenges of using such a blend with contemporary swirling-flow technologies deployed in commercial systems. Finally, recommendations derived from the project are presented for future work.

Chemical-kinetic and Experimental Setup

Zero-dimensional chemical equilibrium calculations were performed using GASEQ to determine the effects on adiabatic temperature and emissions at various pressures and inlet temperatures using the 50:50 blend. As previously observed [22], GASEQ has provided adequate results for NO_x emissions using ammonia blends, thus enabling good correlation with experiments and allowing guidance on the selection of equivalence ratios. The principle of the programme is to balance chemical calculations [28]. The products calculation is performed through the resolution of the free energy equation,

$$\frac{G}{RT} = \sum_{i=1}^{nSp} \left(\frac{x_i G_i^0}{RT} + x_i \ln \frac{x_i}{\sum x_i} + x_i \ln p \right) \quad (1)$$

The blend was then analysed in terms of its Wobbe Index, equation (2), where the higher heating value (HHV) and the specific gravity (S_g) are correlated. The results were compared to other gaseous fuels, Figure 1. As observed, the Wobbe indexes of the ammonia blends fall very close to that of town gas (i.e. 31%CH₄, 49%H₂ and 20%CO₂) and likely similarly to other syngases. Also, it is evident that the increase of hydrogen (that has gone from 0% to 75% in the comparison) does not show a greater increase in the Wobbe Index, a point to be considered when developing combustion systems for these blends.

$$W_o = \frac{HHV}{\sqrt{S_g}} \quad (2)$$

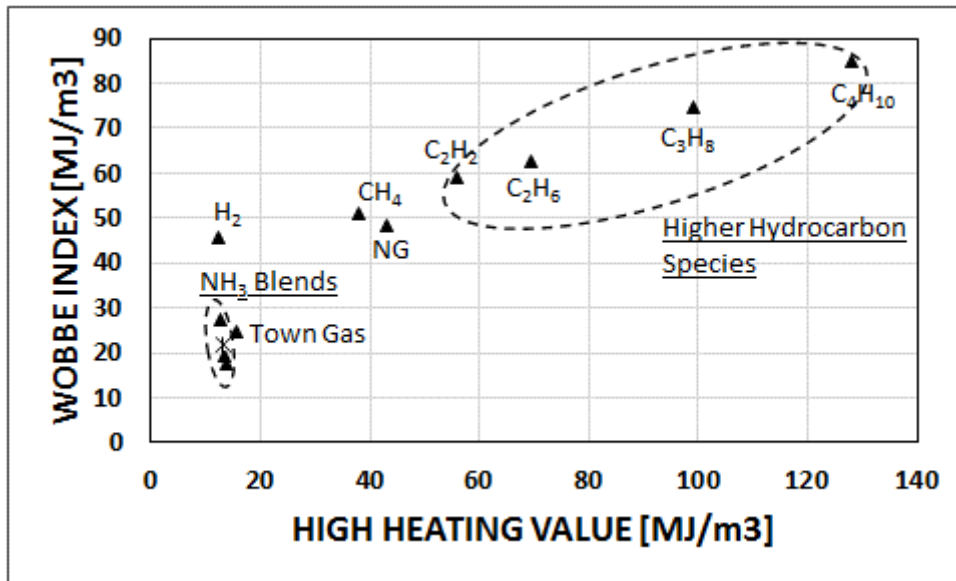


Figure 1. Wobbe index of ammonia blends compared to other traditional gaseous fuels. 50:50 NH_3/H_2 blend is represented by an asterisk.

CHEMKIN-PRO was also used to determine the flame speed of the blend compared to methane, as this is also a critical parameter for the development of stable combustion systems. The analysis was carried out using 5 different reaction mechanisms, selecting those that show the most promising results for the study of ammonia/hydrogen gas turbines as done by Xiao et al. [29]. A reduced model of Konnov's mechanism [23], Tian's [30], Mathieu's [31], Klippenstein's [32], and Miller's [33] were used for the study, Figure 2. The results demonstrate that the flame speed of a 50:50 ammonia/hydrogen blend is close to that produced by methane [34-35] under lean conditions especially using Tian's model. As demonstrated by Hua et al [29], Tian's model is relatively accurate at lean conditions compared to other models, thus providing evidence of the higher flame speed of ammonia while being doped at these hydrogen concentrations, ensuring a responsive flame during the test campaign.

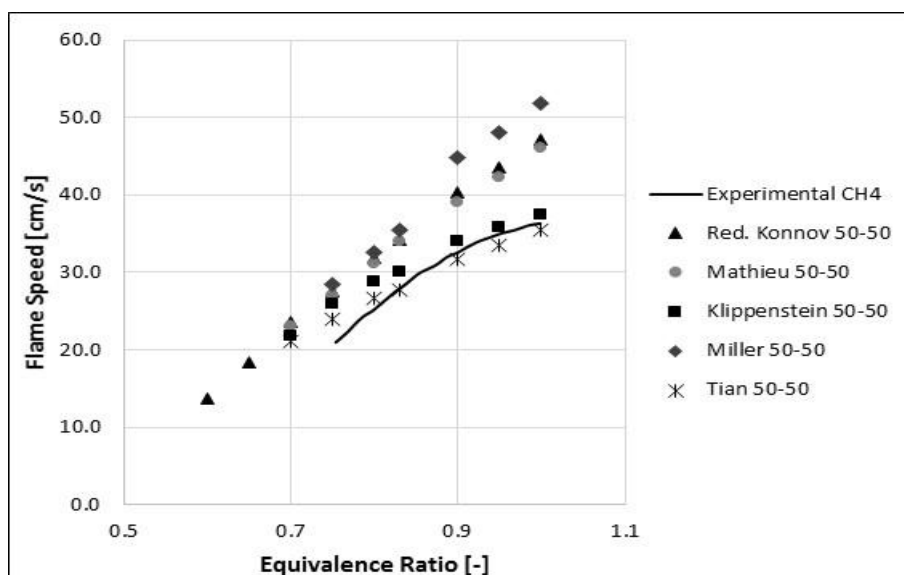


Figure 2. Flame speed analysis using 5 different reaction models.

Tian's model, showing the closest results to methane and best correlation to experiments at lean conditions [29], was used for further analyses on the consumption of species at the flame. Simulation were conducted using both a Laminar Flame Speed calculator and a Chemical Reactor Network (CRN) model that included two clusters formed by a hybrid Perfectly Stirred Reactor-Plug Flow Reactor (PSR-PFR), Figure 3. The initial calculator was used to determine the consumption of species through the flame. For the CRN, the first cluster represents the swirling flame with a central recirculation zone (CRZ) whose recirculation was set at 20% of the product gases. Recirculation strength was approximated from previous experimental campaigns [36].

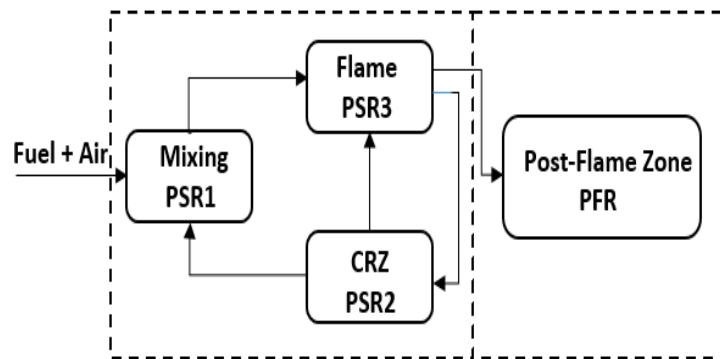


Figure 3. Gas turbine network for 1-D modelling.

Burner experiments were performed at the Gas Turbine Research Centre (GTRC) of Cardiff University. A generic swirl burner developed in-house was used for this purpose. The rig has been used in a great variety of experimental campaigns to assess new alternative fuels in gas turbine combustion [37]. The swirl chamber is fitted to the premixing chamber, thus allowing greater residence time for mixing and increasing pressure, which reduces the likelihood of flashback and overheating. An interchangeable tangential swirler attaches to the premixing chamber. This swirler has different nozzle geometries at different angles and can be modified easily, thus giving both different swirl numbers and outlet conditions. The entire rig is confined by a quartz tube fitted to the system to provide a confinement for the flame, as utilised in modern gas turbine combustors, Figure 4. Coriolis mass flow meters are used and standard to achieve precise measurement of flowrates with an accuracy of $\pm 0.5\%$ RD plus $\pm 0.1\%$ FS. Further information can be found in previous works [22, 37].

Various equivalence ratios were investigated using a geometric swirl number of 1.05. Re varied from 10,800 to 21,000 for a range of operating conditions during the experimental programme. An inlet temperature of 288K and atmospheric pressure were maintained during the tests. A 31.5 kW power load was studied under conditions specified in Table 1. An Avigilon 5 Mp H.264 HD camera was used to capture the flame during the various stability stages and provide qualitative insights of the flame behaviour.

In order to stabilise the flame initially, a stable methane flame was achieved prior to the injection of hydrogen. Hydrogen was then injected until a stable methane/hydrogen flame was achieved at ~ 30 kW net thermal power. Once stabilization was achieved, ammonia was

slowly injected while reducing methane simultaneously, until a 50:50 hydrogen/ammonia blend was achieved.

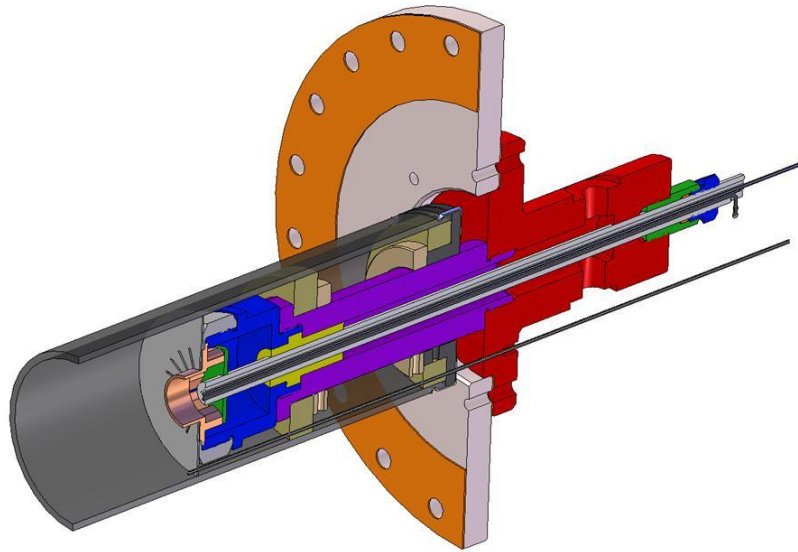


Figure 4. Generic Swirl Burner.

Table 1. Experimental Matrix at 50% NH₃ and 50% H₂.

NH ₃ [g/s]	H ₂ [g/s]	O ₂ [g/s]	Air [g/s]	Power [kW]	ϕ
0.80	0.09	4.43	19.02	31.35	0.41
0.80	0.09	4.22	18.10	31.35	0.43
0.80	0.09	4.00	17.16	31.35	0.46
0.80	0.09	3.50	15.04	31.35	0.52
0.80	0.09	3.26	14.00	31.35	0.56

A heated vacuum chemiluminescence analyser (Signal 4000VM) was employed to quantify NO_x concentrations, calibrated to 37.1 ppm_v NO and 1.9 ppm NO₂. NO_x results showed fluctuations of $\pm 4.0\%$ during the trials. The system also contains a multi-gas analyser that can not only measure CO and CO₂ (not used in this study) but also includes a paramagnetic sensor for O₂ readings (up to 22.5%mol), with a measured error of $\sim 1.0\%$. As a backup and for data confidence, a standalone Rosemount NGA 2000 multi-gas analyser was employed, which provides secondary readings of O₂, all calibrated to the same concentrations previously stated with a measured error $\sim 1.5\%$.

Results

O-D GASEQ Analysis

Initial results were obtained through 0-D Gaseq analyses to determine the reduction of NO_x emissions using a 50:50 blend, as presented in Figure 5. It must be emphasized that these calculations consider combustion equilibrium of chemical species, whilst ignoring the considerable impact of the swirling flow hydrodynamics and complex heat transfer phenomena, thus can only be used as a starting reference point.

It is clear from Figure 5 that the use of a lean premixed blend $\phi \sim 0.5$ almost halves the production of NO_x to the one produced at $\phi \sim 0.8$.

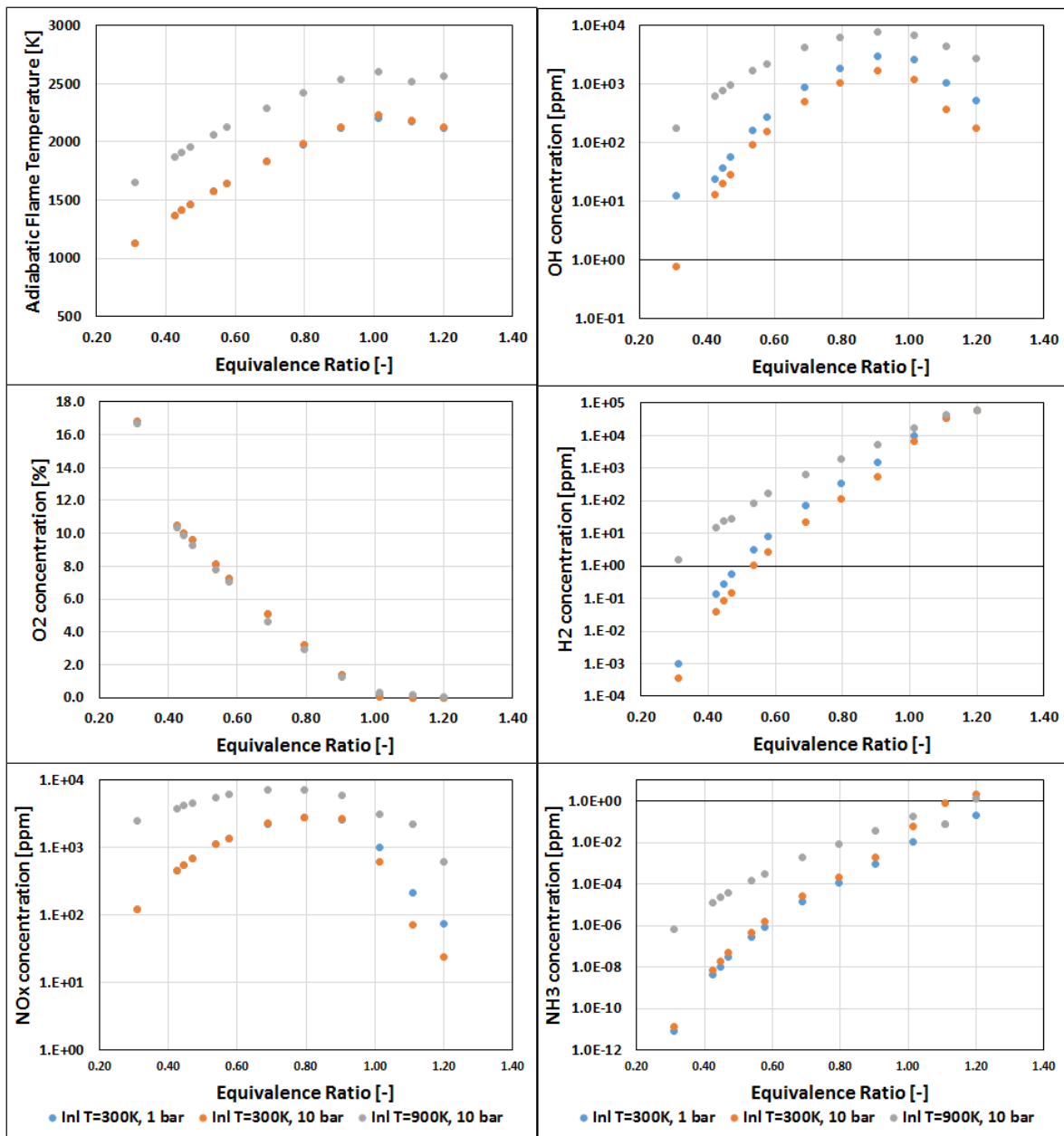


Figure 5. Flue gases profiles at various inlet temperatures and pressures.

The results also indicate that ambient pressure appears to only have a minor effect on the emissions profile, with just a minimum impact on the production of NO_x . By contrast, the inlet temperature has a considerable effect on emissions, especially NO_x , NH_3 and OH, with a clear increase in adiabatic temperature of ~ 300 - 450 K. This increase in emissions is related to the reaction of OH radicals, as seen later, as well as the Zeldovich phenomena. As the inlet temperature has increased, also the amount of OH and O due to the higher reactivity of the reactants O_2 and H_2 . This increase will allow further interactions with the hot ammonia with a concentration decrease of the species in ~ 5 (O). High combustion temperatures will also produce further NO_x via the Zeldovich mechanism, considerably increasing emissions in the flue gas stream.

1-D Reactivity analyses

Reaction analyses were carried out using a 1-D model to determine the reactivity of the different species through the flame, Figure 6. The results at $\phi=0.52$ show that the consumption of hydrogen occurs first consequence of the highest reactivity and diffusivity of the molecule, as expected. In order to observe the impact of equivalence ratios on the reactants, a parallel calculation was performed at $\phi=0.8$, Figure 6b. It must be noticed that the molar fraction of species has increased due to the reduced amount of air in the system, compare Figures 6a and 6b. The results show how the reaction of hydrogen increases at leaner fuel conditions, with the molecule reacting through a flame envelope of $\sim 0.6\text{cm}$ compared to only $\sim 0.2\text{cm}$ under richer (i.e. $\phi=0.8$) conditions.

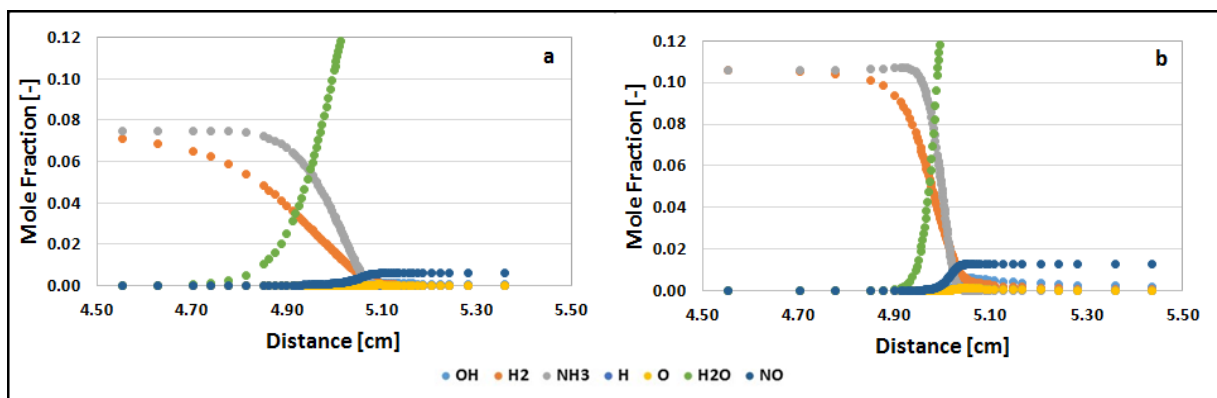


Figure 6. 1-D Model for 50:50 ammonia/hydrogen reaction at a) $\phi=0.52$; b) $\phi=0.80$.

CRN Reaction analyses

Further analyses were conducted to show the rate of production and consumption of radicals at $\phi=0.52$ while immersed in a recirculating flow closer to those observed in swirling flows, Figure 7. The results denote the consumption of hydrogen and ammonia through their reaction with OH, O and H. Thus, the excess of OH radicals under these lean conditions enhance the production of other radicals such as NH_2 which can then lead to the formation of HNO, molecule that precedes the formation of NO.

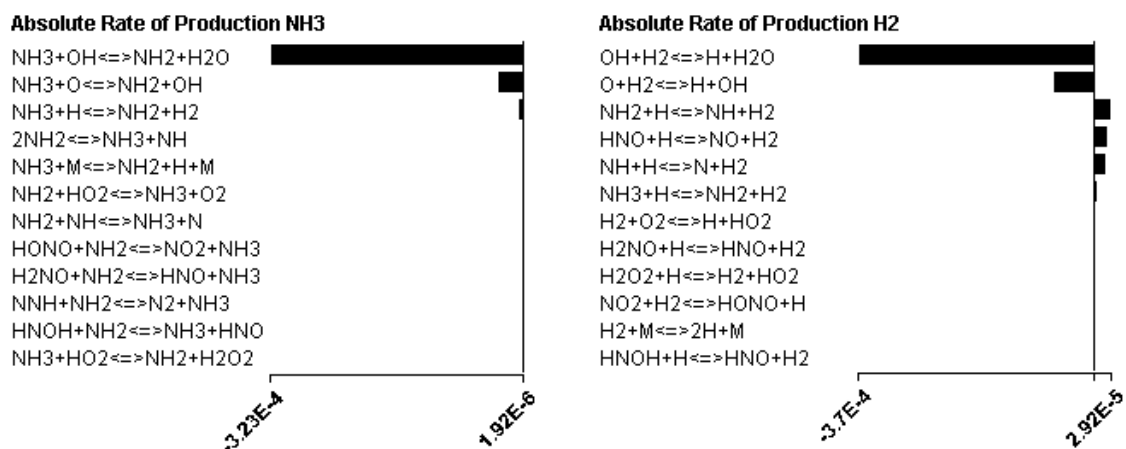


Figure 7. Absolute Rate of Production of NH_3 and H_2 in a recirculating flow, respectively.

Although the adiabatic flame temperature at lean conditions is $\sim 1000\text{K}$ lower than at stoichiometric conditions, Figure 5, the high nitrogen concentration in the ammonia molecule seems to react with the OH radicals formed in excess due to the high oxygen flowrate. Therefore, OH radicals in the flue gases - which have a low concentration in Figure 5 as they have already been consumed - not only produce NH_2 that reacts to form NO via the reaction $\text{NH}_2 + \text{O} \rightarrow \text{HNO} + \text{H}$, Figure 8, but also react with the nitrogen through the reaction $\text{N} + \text{OH} \rightarrow \text{NO} + \text{H}$, as stipulated by Li et al [36]. Thus, the increase of NO under lean conditions is 1(O) greater than under rich fuel conditions.

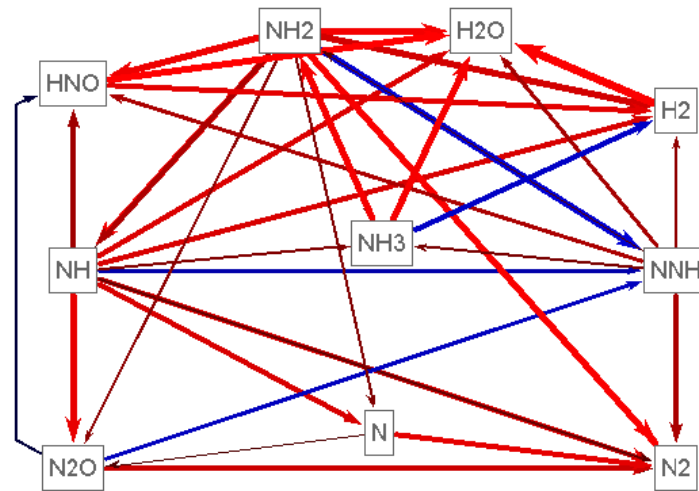


Figure 8. Ammonia reaction path. NHO is a large by-product of NH_2 at these lean conditions.

Experimental campaign

An experimental study was undertaken in order to assess the use of this blend on the production of NO_x and the stability of combustion under lean premixed conditions. NO_x emissions were very high - in the range of thousands of ppm for equivalence ratio $\phi > 0.5$, Figure 9. The results are in good agreement and of the same order of magnitude as for the 0-D calculations (Figure 5). As discussed earlier, the high OH and O concentrations are the main problem for the formation of nitrogen oxides under these conditions.

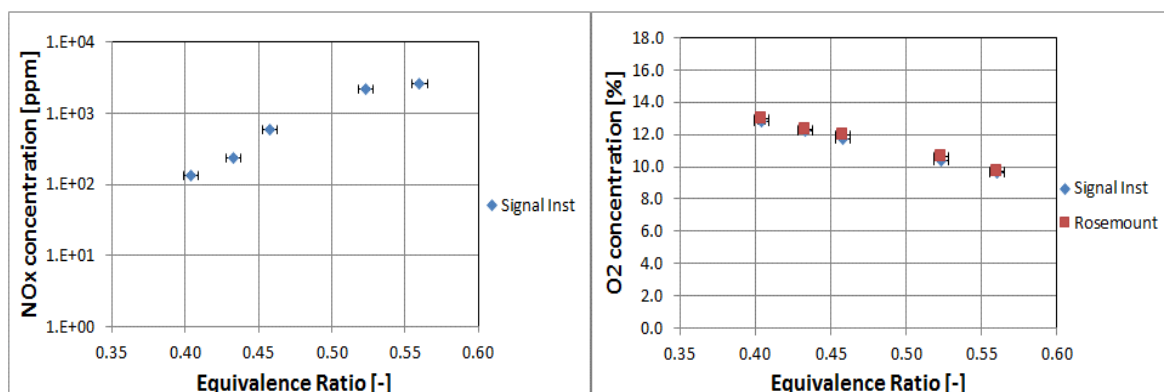


Figure 9. Experimental NO_x emissions and oxygen concentration in the flue gases for a 50:50 ammonia/hydrogen flame. Inlet temperature 288K, atmospheric pressure.

In terms of combustion stability, it was observed that above an $\phi = 0.43$, the flame was stable, Figure 10. It was notable that the hydrogen seemed to be burning first, as a consequence of

its higher diffusivity and reactivity, as previously seen in Figure 6. Qualitative evidence of this phenomenon is seen close to the burner mouth, where a thin, almost invisible layer that surrounds the inner flow appears, i.e. as hydrogen flames are well-known to be almost invisible, while ammonia burns with an orange colour with intense spectra at 543.6, 571.3, 604.2 and 665.2 nm product of NH_2 reaction [39]. This assumption correlates to the previous 1-D analysis.

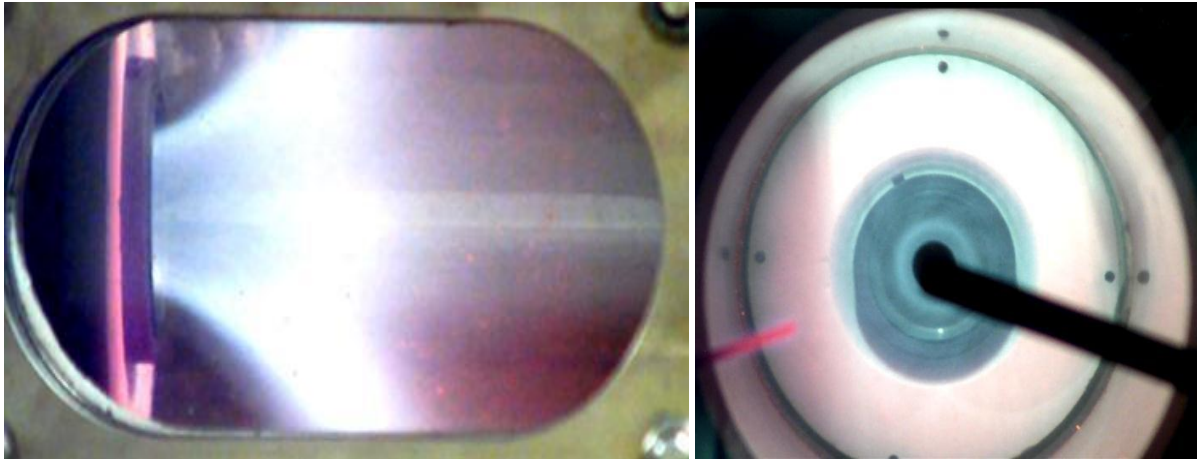


Figure 10. Flame with an ammonia-hydrogen blend. Equivalence ratio 0.43ϕ . Left) Axial-tangential plane; right) radial-tangential plane. Flame $\sim 0.45\text{m}$ in length.

Once the flame had stabilised, its emission performance was better than methane-ammonia blends studied previously [22]. However, just above blowoff conditions, with equivalence ratios at about 0.50, the flame presented an unstable, pulsating nature with traces of flame re-ignition produced by the hydrogen at frequencies between 10-15Hz.

At higher equivalence ratios $\phi \sim 0.52$, Figure 11, the flame started vibrating with less stability. The hydrogen started to burn before the ammonia, with the hydrogen flame sitting close to the burner tip. It was also evident that the establishment of the hydrogen flame near the boundaries of the nozzle was the phenomenon that precedes boundary layer flashback [40] as a consequence of the low Re numbers imposed at these power outputs.

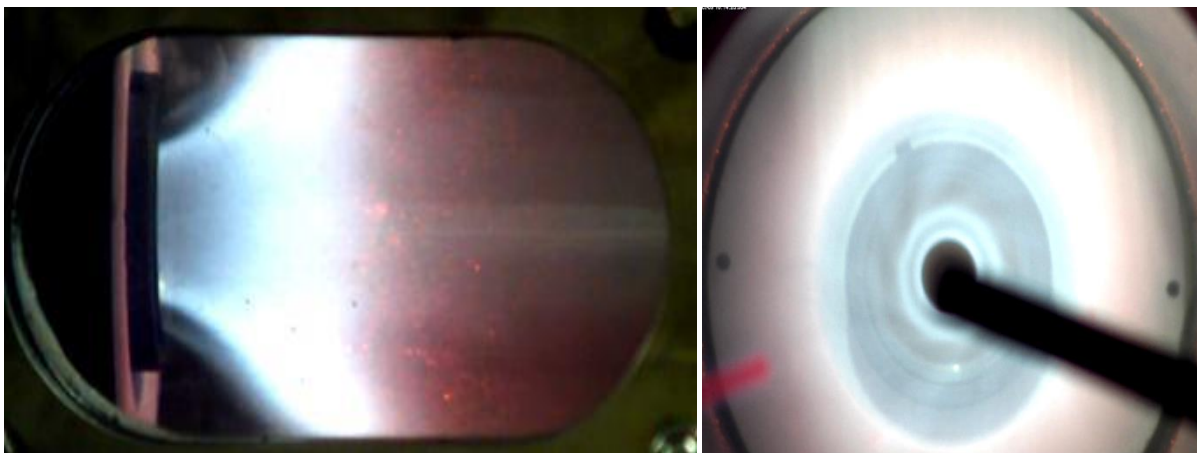


Figure 11. Flame with an ammonia-hydrogen blend. Equivalence ratio 0.52ϕ . Left) Axial-tangential plane; right) radial-tangential plane. Flame $\sim 0.42\text{m}$ in length.

A whiter flame was observed downstream, showing traces of low combustion efficiency at the tip of the flame. It is emphasized that optical access to the flame was performed volumetrically rather than using planar non-intrusive techniques, thus adding thickness to the tip of the flame. However, the whiter colour, compared to a pure ammonia flame, has been correlated to the increased consumption of OH and O radicals.

Recommendations

The results show promising data for future research and designs. A novel combustion system could be developed with a combination of gaseous streams where hydrogen could be burned prior to ammonia to aid the decomposition of the NH_3 molecule, whilst allowing more oxygen to get in contact with the already split NH_3 (i.e. NH_2 radicals). The high hydrogen concentration downstream of the combustion zone could be recirculated, allowing further combustion of radicals with the remaining hot oxygen. Moreover, recirculation of flue gases could enhance still further the reduction of NO_x emissions without compromising the initial hydrogen flame. It is proposed that the use of swirl can benefit these hybrid flames significantly, but not at the values deployed in this study. As observed and known from other experiments [41], high swirling conditions increase flashback propensity due to the impact of enhanced coherent structures, particularly the Central Recirculation Zone. Thus, reduced swirl numbers can benefit this particular blend by a) allowing enough time for the ammonia to react in the combustion chamber, as this will improve combustion [42]; b) hydrogen can decrease its interaction with the oxygen close to the tip of the burner; c) reduce the strength of detrimental coherent structures that can lead to early flashback. Finally, due to its low Wobbe index, it is recommended that initial designs are based on current combustion systems based on syngas/town gas applications, as interchangeability with these systems would be easier.

Conclusions

Research was carried out to determine the combustion features and emission profiles of a 50:50 hydrogen-ammonia blend. It was recognised that the NO_x are still high compared to rich conditions as a consequence of the excessive production of OH and O radicals that interact with the ammonia, hydrogen and nitrogen from the fuel and/or the air. Moreover, it was observed that the flames produced using this blend can be stabilised at equivalence ratios $0.43 < \phi < 0.52$ for a thermal net power of $\sim 31\text{kW}$. This operability range is considered to be narrow compared to those associated with other fuels. This is caused by the high diffusivity and reactivity of hydrogen combined with the high swirl number that finds the oxygen of the blend and starts reacting, thus producing boundary layer flashback due to the low Re numbers imposed at these powers. However, the high production of hydrogen, oxygen and OH radicals under these conditions show promising results for further design of low-swirl combustors with stratified hydrogen injection for stable operation.

Acknowledgements

The authors gratefully acknowledge the support of Siemens AG through project 507698. The authors would like to thank Mr Steve Morris for his inputs, suggestions and GTRC facility management.

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