

Experimental and simulation investigations of the combustion of blends of ammonia with hydrogen and natural gas in industrial non-premixed burners

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Abstract

In addition to hydrogen, ammonia is an interesting energy carrier for a globally decarbonized energy system. The combustion of ammonia is not without challenges, however: it is far less reactive than natural gas or hydrogen, with consequences for combustion stability, while the emissions of nitrogen oxides are likely to be higher. One potential application of the combustion of ammonia or ammonia blends could be in industrial manufacturing processes. They usually employ non-premixed combustion to generate process heat, which may be advantageous in the context of a difficult fuel such as ammonia. Some of the first investigations on industrial combustion processes are presented, using both ammonia and ammonia blends with either hydrogen or natural gas, based on measurements with industrial burners in GWI's semi-industrial combustion test rigs and the accompanying analyses using chemical kinetics.

Introduction

In the context of global decarbonization efforts, industrial process heating is also a relevant topic. In Germany, more than 20 % of the country's final energy consumption is due to heat generation in industrial manufacturing processes, for the most part by burning natural gas or other fossil fuels. Decarbonizing process heat is a significant challenge, due to the high degree of heterogeneity and specialization in the various industries. While electrification is one option, especially for low-temperature process heat, high-temperature processes, e. g. in the metals, glass or ceramics industries, may still require some kind of combustion process. Hydrogen (H_2) is often considered the premier choice for a carbon-free fuel, but brings its own challenges due to its low volumetric calorific value and extremely low boiling temperature (about $-250\text{ }^\circ\text{C}$), among other things. While gaseous hydrogen can be transported in pipelines, transporting or storing it in tanks in either liquefied or compressed form is much more difficult.

Another interesting energy carrier is ammonia (NH_3) which is much easier to transport (boiling temperature: $-33\text{ }^\circ\text{C}$ in standard conditions), making it particularly interesting for supplying users without a connection to a decarbonized energy infrastructure. NH_3 could be cracked on-site to produce H_2 as a fuel, but it could also be burned directly to avoid conversion losses. However, the use of NH_3 as a fuel poses some challenges, for example in the context of flame stability and pollutant emissions.

Ammonia as a fuel for industrial process heating

Compared to more conventional fuels such as natural gas or even hydrogen, burning ammonia can be somewhat difficult: while the calorific value of ammonia is higher than that of hydrogen. ($H_i(NH_3) = 14.14\text{ MJ/m}^3$; $H_i(H_2) = 10.8\text{ MJ/m}^3$; $H_i(CH_4) = 35.89\text{ MJ/m}^3$, all values given in $25\text{ }^\circ\text{C}/0\text{ }^\circ\text{C}$, 1 atm), its adiabatic combustion

temperature is significantly lower ($1,800\text{ }^\circ\text{C}$ vs. $1,950\text{ }^\circ\text{C}$ and $2,100\text{ }^\circ\text{C}$ for methane and hydrogen respectively). Ignition and combustion stability are an issue, given the very high minimum ignition energy of ammonia [1], but extremely low laminar combustion velocity, compared to H_2 or CH_4 (cf. **Figure 1**).

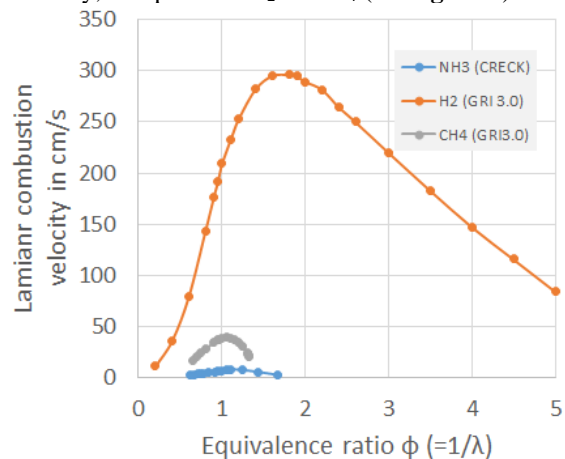


Figure 1: Laminar combustion velocities ($p = 1\text{ bar}$, $T_{initial} = 300\text{ K}$) of NH_3 , CH_4 and H_2 . [2] was used for the calculation with NH_3 , [3] for the calculations for CH_4 and H_2 .

Another combustion-related aspect is the propensity of ammonia to produce high concentrations of nitrogen oxides (NO_x). While the thermal formation mechanism usually is the most dominant route for NO_x formation in conventional gaseous fuels, particularly in high temperature applications, NO_x formation due to fuel-bound nitrogen is likely to play a much bigger role when burning ammonia [4]. Given the strict emission limits imposed on industrial manufacturing processes, complying with these limits when using NH_3 will be challenging, even considering the way that pollutant emissions are affected by flue gas sample drying when burning hydrogen-rich fuels which can distort comparability with emission levels from more conventional fuels such as natural gas [5].

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In contrast to gas-fired applications in the residential or power sector, combustion systems used for industrial process heating are usually non-premixed, in order to facilitate air preheating or oxy-fuel combustion [5], [6]. Therefore, the mixing of fuel and oxidizer is crucial for the performance of industrial burners. The predominance of non-premixed combustion in industrial process heating may also offer some opportunities in the context of difficult fuels, as this form of combustion generally allows for much better stability than the premixed combustion that can be found for example in gas turbines.

As ammonia is a relatively new fuel for industrial process heating, there are still many open questions about its combustion behavior and how to best address its specific challenges. Therefore, GWI has started to carry out initial investigations on how to use NH₃, using chemical kinetics, preliminary CFD and experiments both on a workbench and semi-industrial scale (firing rate 180 kW).

Ammonia as a fuel for industrial process heating

As mentioned above, the use of NH₃ as a fuel has proven to be a non-trivial operation even in the first experimental investigations on a semi-industrial scale. To investigate commercial burner systems using ammonia, initial tests were carried out at GWI. The burner used is a non-premixed burner with a ceramic flame tube (SiC), type BIC, manufactured by Honeywell, and is designed to burn natural gas at a nominal firing rate of 180 kW. It was operated in a high temperature test rig at a furnace temperature of approximately 875 °C. In order to compensate for the unfavorable combustion characteristics of ammonia in terms of flame stability, initial tests were carried out with auxiliary gases consisting of natural gas (NG) and hydrogen which were blended with the ammonia. The successive blends allowed to explore the limits of flame stability during the tests.

The ammonia fuel fraction value is defined by Equation (1):

$$x_{NH_3} = \frac{V_{NH_3}}{V_{NH_3} + V_{support\ gas}} \quad (1)$$

The *Figure 2* shows that the combustion limit of an LNG-NH₃ gas mixture already leads to flame extinction at a low admixture rate (7.25% by volume NH₃). In view of this, the tests were repeated at partial load (60% load) and flame stability was maintained up to a volumetric NH₃ content of 60%. In terms of flame stability, blending with hydrogen proved to be significantly more stable. Blending rates of 65% (full load) to 70% (60% load) ammonia in hydrogen were achieved. Based on this, it can already be seen that, in addition to the combustion properties of the NH₃-NG and NH₃-H₂ fuel gas mixtures, the local flow conditions and burner geometry also play an important role in the flame stability of NH₃ flames. In addition to flame stability, the experimental investigations also looked

into the formation of pollutants. The focus was on the resulting NO_x concentration in the flue gas. In order to ensure sufficient comparability of the resulting NO_x concentrations when using different fuel gas compositions, it is useful to select a suitable reference. Therefore, the following evaluations of NO_x concentrations are related to mg / kWh. Equation (1) shows the conversion of the measured NO_x value in [ppm] to the equivalent value in [mg/kWh] based on EN 267 [7]:

$$NO_x = NO_{x,meas} \cdot 2.056 \cdot \left(\frac{21}{21 - O_{2,meas}} \right) \cdot \left(\frac{V_{A,th,tr,min}}{H_i} \right) \quad (2)$$

In the context of the energetic use of ammonia, the concentrations of N₂O and NH₃ also play an important role. N₂O is a greenhouse gas with a global warming potential (GWP for 100 years) of 265 according to IPCC Climate Report [8]. Ammonia slip must always be avoided due to its odor and toxicity.

As can be seen in the diagram, the resulting NO_x concentrations of the respective ammonia blends differ significantly in both height and curve shape. The NO_x formation for the NH₃-H₂ mixture shows a parabolic curve with a maximum value of approx. 5,000 mg/kWh. With increasing NH₃ content, the NO_x concentration decreases rapidly.

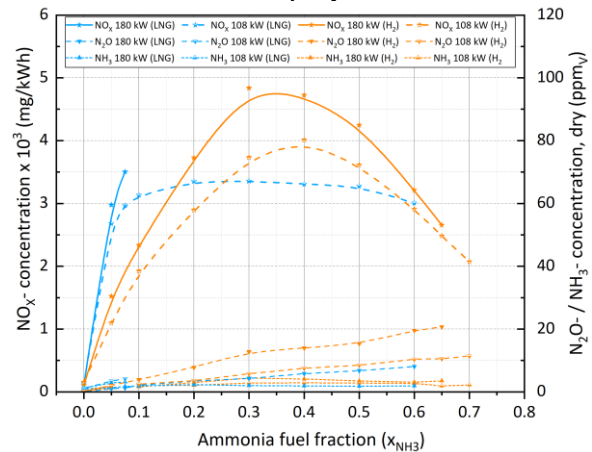


Figure 2: NO_x-concentration depending on ammonia fuel fraction; Combustion of Ammonia blends with LNG and Hydrogen while using an industrial burner system.

In addition to the NO_x curve mentioned above, a relatively linear correlation of the N₂O emission with the NH₃ content in the fuel gas was also found in the measurement series. In contrast, only small amounts (< 5 ppm_v, dry) of unburnt ammonia were detected in the flue gas during the tests, even at high admixture rates. The combustion of ammonia blends based on LNG shows a different curve of the NO_x concentration depending on the NH₃ content in the fuel gas. Even small amounts (x_{NH₃} = 0.05) lead to a drastic increase of NO_x compared to the value of pure LNG combustion. In contrast to the NH₃-H₂ blends already mentioned, the curve develops a plateau from an NH₃ content of approx. 10 %. The tendency towards a decreasing NO_x concentration, which became apparent

at the highest admixture rates, could not be further investigated due to the flame instability.

The works of Alkhateeb et. al. [9] provide an interesting trend for estimating the NO_x emissions at higher admixture rates. In the tests with a premixed burner, $\text{CH}_4\text{-NH}_3$ flames were examined. From an NH_3 content of 80 %, the NO_x concentration, which is high per se, decreases as the NH_3 content increases. In addition to the reducing effect of high NH_3 proportions in the methane mixture, the diagram (see also *Figure 7*) shows a strong dependence on the equivalence ratio. With this in mind, the equivalence ratio was varied in addition to the blending rates. The following *Figure 3* shows the NO_x curves for NG and hydrogen combustion as well as the gas mixtures from *Figure 2*. The NO_x curves of NG and hydrogen combustion show an analogous linear decrease of the NO_x concentration in the direction of stoichiometry and differ only slightly in the absolute concentration. The conversion to [mg/kWh] mentioned above takes into account the resulting different stoichiometric flue gas volumes (see *equation 2*).

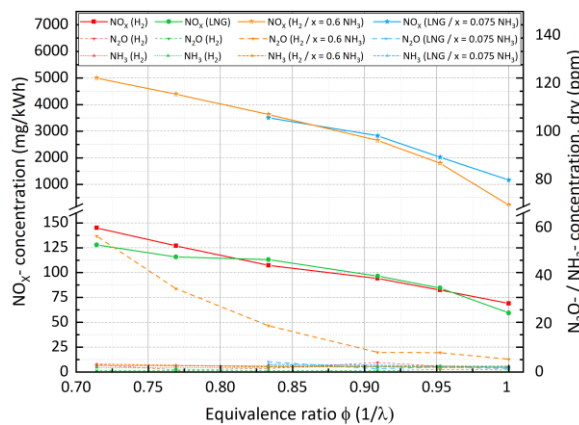


Figure 3: NO_x -concentration depending on equivalence ratio; Combustion of Ammonia blends with NG and Hydrogen, NG and Hydrogen while using an industrial burner system.

Figure 3 also shows a clear difference between the NO_x formation of the auxiliary gases (NG, H_2) and the blended combustion with ammonia. The reason for this can be found in the NO_x formation pathways. While the thermal NO formation mechanism (Zeldovich mechanism) dominates in the case of NH_3 -free fuel gases, the fuel NO mechanism is also important in the case of ammonia-containing fuel gases. In contrast to thermal NO , fuel NO is preferentially formed from nitrogen compounds in the fuel gas molecules (NH_3). According to Warnatz [10], much of this fuel-bound nitrogen is oxidised to nitrogen monoxide during combustion under lean conditions.

As already mentioned, the absolute NO_x concentrations for the NH_3 -containing fuel gases are far higher than those for the NH_3 -free fuel gases. Regardless, all the NO_x curves in *Figure 3* show the same qualitative behaviour. The highest NO_x concentrations for all fuel gases are found at the leanest settings. The lowest are

in the near stoichiometric mode. The absolute reduction is considerable, especially for the NH_3 -containing fuel gases: as can be seen in *Figure 3*, the NO_x concentration for LNG/ammonia combustion drops by 66% just by increasing the equivalence ratio from 0.7 to 1. An even more significant effect of the air reduction can be seen in the combustion of the H_2 -ammonia fuel gas: here the NO_x concentration was reduced by > 95%. Similar to the NO_x curve already mentioned, the N_2O curve shows an initially high N_2O concentration at leanest conditions and decreases with increasing equivalence ratio. On the other hand, none of the test series show an NH_3 concentration above 3 ppmv.

In conclusion, the first experimental investigations with an industrial burner showed a tendency to high NO_x concentrations during the combustion of ammonia mixtures with both natural gas and hydrogen. However, the measures observed to reduce the high NO_x levels, such as increasing the NH_3 content in the fuel gas mixture and near-stoichiometric combustion, are positive.

Additional laboratory experimental studies

Based on these first findings, additional experimental investigations were carried out on a laboratory test rig to investigate higher NH_3 admixture rates and even pure NH_3 combustion. The non-premixed burner used is a GWI proprietary development and was manufactured by the company Kueppers Solution using a 3D printing process (Laser Surface Melting, LSM). *Figure 4* shows the burner-combustion chamber combination and the burner in section.

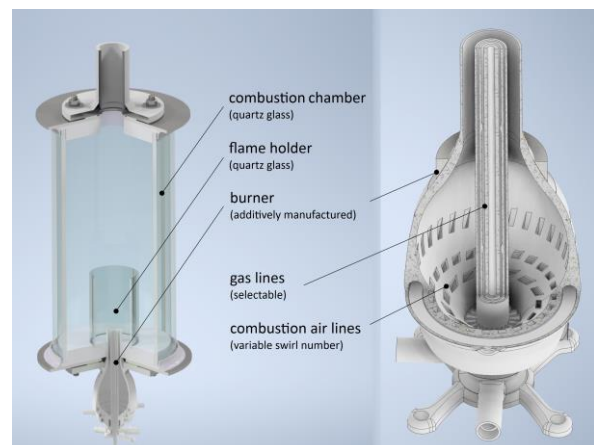


Figure 4: Modell of the burner design including the mounting at the combustion chamber test rig.

In view of the unfavourable combustion properties of ammonia, a geometric variability of the swirl flow of the combustion air was implemented in the burner design. The aim was to investigate the flame stability limits with respect to both the maximum ammonia admixture rate and the swirl number.

The swirl number is, according to Leukel [11] a dimensionless value for characterising swirl flows and is calculated from the ratio of the angular momentum flow to the product of the axial momentum flow and the

reference radius (see *equation 3*). The proportion of the angular momentum flow is primary determined by the design, e.g., the angle of the blades of a swirler.

$$S = \frac{\dot{D}}{\dot{I} \cdot R_{pr}} \quad (3)$$

In that context, the used burner provides a special feature: during operation, swirl numbers of 0.3, 0.5 and 1.1 can be achieved by manually switching the respective flow channels.

In addition to the three swirl levels, it is also possible to direct the air into the combustion chamber without any tangential flow components ($S = 0$).

As the previous experimental investigations have shown, fuel gas blends consisting of natural gas (NG) and ammonia are of particular interest regarding flame stability behaviour. For this reason, the remainder of this paper is limited to the results of the experimental investigation of $\text{CH}_4\text{-NH}_3\text{-air}$ flames. The following part shows the first results of the laboratory experimental studies.

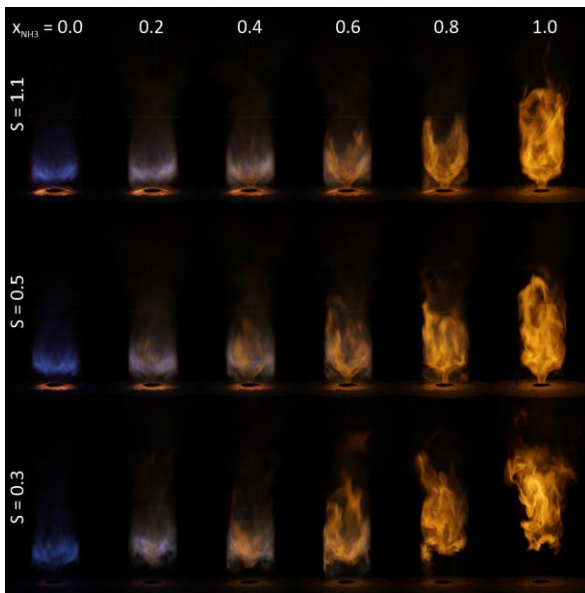


Figure 5: Images of ammonia-methane-air flames for different ammonia contents and different swirl numbers.

Figure 5 shows images of the resulting flames of $\text{CH}_4\text{-NH}_3$ fuel gas mixtures in the range $0 \leq x_{\text{NH}_3} \leq 1$ in 0.2 steps. As can be seen, pure ammonia combustion can be achieved with this burner system. The equivalence ratio is constant at $\Phi = 0.95$ in all the series of measurements shown. In addition to the NH_3 content in the methane, the swirl number was also varied.

The attempt to operate the burner without swirl ($S = 0$) failed already at the operating point with pure methane. An explanation for this can be found in the bottom row of Figure 5. At a swirl number of 0.3 it was possible to stabilise a methane flame, but this is an off-lifted flame that is not suitable, especially with regard to an burner in an industrial furnace.

However, it was also observed that this off-lifted flame shape does not lose its stability even when ammonia is added successively. It is also established in pure ammonia combustion due to the local flow field in the flame tube. When switching to the next higher swirl number ($S = 0.5$), the shape of the flame changes in such a way that it is rooted to the burner mouth can be seen. This flame root remains present up to and including pure NH_3 combustion. However, it is striking that the visible reaction zone expands proportionally as NH_3 is added. The compact reaction volume of pure methane combustion grows in visible volume with increasing NH_3 content. An analogous behaviour can also be seen in the resulting flame shapes of the measurement series at the highest swirl number investigated ($S = 1.1$). By further increasing the swirl number, the flame root becomes even more pronounced and a more compact flame shape can be seen as well. There is also a clear change in the radiation behaviour of the resulting flames in addition to the change in flame geometry due to the increasing addition of ammonia. While the flames without NH_3 show the typical blue colour of a well-mixed methane flame, the addition of ammonia replaces the blue type with a whitish, slightly yellowish flame colour already at a low admixture ($x_{\text{NH}_3} < 0.05$). With increasing ammonia content, the yellowish radiation increases significantly. From an admixture rate of $x_{\text{NH}_3} \geq 0.6$, a dark yellow flame appears, similar to that of a fuel oil flame.

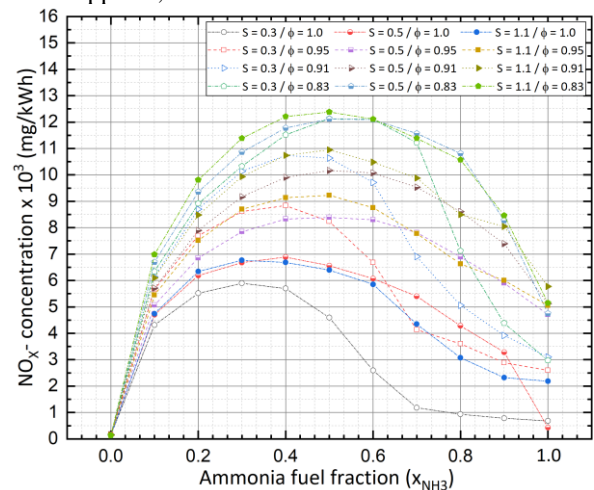


Figure 6: NO_x -concentration in $[\text{mg}/\text{kWh}]$ as a function of ammonia fuel fraction, equivalence ratio, and swirl number.

In addition to investigating the swirl-dependent flame stability and observing the visual changes in the combustion reaction, the tests shown in Figure 5 provided further insight into the NO_x formation behaviour. In principle, when considering the NO_x emission values, the test series with similar flame geometry should be compared with each other. This is the case for the high swirl series ($S = 0.5$ and 1.1). The test series with the lowest swirl deviates significantly from the previous ones in its flame shape.

Figure 6 below shows the concentration curve of NO_x as a function of the ammonia content in the methane fuel gas mixture. The data points shown are the results

of a total of twelve different series of measurements. In addition to the variation of the swirl number in three steps, the equivalence ratio was also varied in a range of $0.83 \leq \phi \leq 1$ in four steps.

The NO_x curves of the tests with swirl numbers 0.5 and 1.1 show a similar behaviour in the form of a clear increase in concentration with a low ammonia admixture rate ($x_{\text{NH}_3} \leq 0.05$) and a parabolic curve with a concentration maximum at an ammonia content of approx. $x_{\text{NH}_3} = 0.5$. The lowest NO_x concentrations in combustion with NH_3 containing fuel gases are observed in pure ammonia combustion. For example, the maximum NO_x concentration is halved when burning pure ammonia with parameters $S = 1.1$ and $\phi = 0.83$.

In addition to the dependence of the NO_x concentration on the degree of NH_3 admixture, other factors already mentioned have an influence on the NO_x reduction. One effective method was also confirmed in these tests: near-stoichiometric combustion showed the lowest NO_x concentration values of the respective measurement series for each of the swirl numbers investigated. The following Figure 7 illustrates this effect and provides a comparison with literature data. It should be noted that this is a comparison with a premixed burner. In principle, only tendencies can be compared and not absolute values. Figure 7 shows the results of a series of experiments by Alkhateeb et. al. [9]. The diagram has been extended to include these measurement points in order to classify the results obtained with the GWI burner. Only the $S = 1.1$ and 0.5 series were included in the diagram for the reasons given above. The measured data were referenced to a residual oxygen concentration of 6 vol% in the dry flue gas according to the axis labelling. Considering the forms of combustion (e. g. premix, non-premixed), the results fit well with the literature data. In this respect, apart from the tendency for the concentration to decrease as the equivalence ratio increases, the level of the measured NO_x concentration does not appear to deviate excessively.

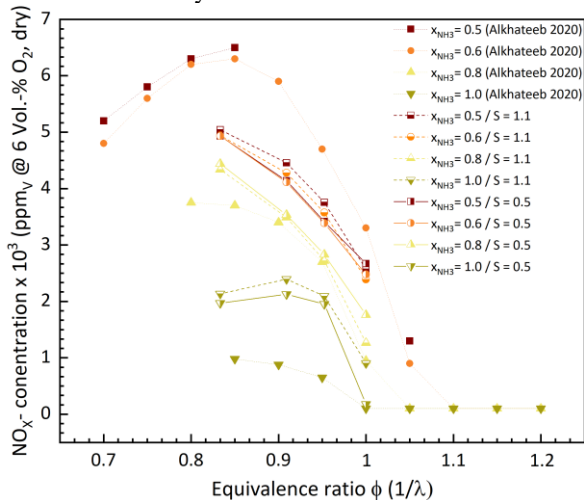


Figure 7: Measured exhaust NO_x -concentration in parts per million (ppm) as a function of equivalence ratio. Comparison with Alkhateeb et. al. [9]

It is also clear that, in addition to the influences already mentioned, the formation of NO_x is also dependent on the shape of the flame itself. The series of measurements at a swirl number of $S = 0.3$ provides an important example (see Figure 6). In contrast to the previously described series, this one shows a NO_x maximum at approximately $x_{\text{NH}_3} = 0.5$, which is dependent on the equivalence ratio. The maximum is observed with increasing equivalence ratio at lower NH_3 addition rates (0.6 / 0.4 / 0.3). There is also a change in the parabolic course of the NO_x concentration: after the respective maximum values, the concentration decreases in a degressive course. The beginning of the degressive curve corresponds well with the visible change of the flame geometry at $x_{\text{NH}_3} = 0.6$. From this concentration, the flame is more wavering and not fixed in its position. In general, however, especially in the series of measurements with $\phi = 1$, the swirl number 0.3 has the lowest mean NO_x concentration.

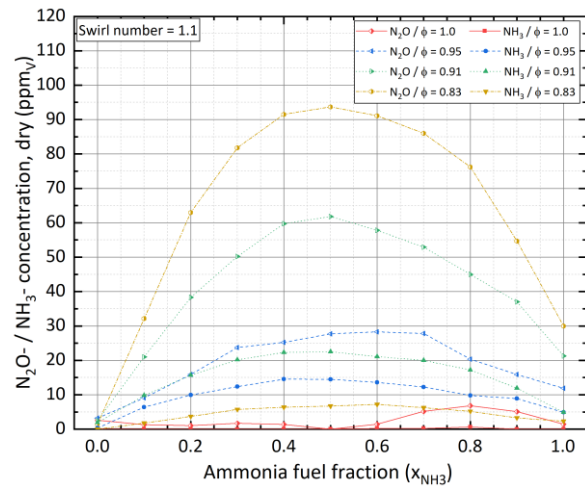


Figure 8: Measured exhaust NH_3 and N_2O concentrations in parts per million (ppmv) as a function of ammonia fuel fraction and equivalence ratio; Swirl number: 1.1.

In addition to the maximum high NO_x concentration in the average ammonia admixture range in methane ($0.4 \leq x_{\text{NH}_3} \leq 0.6$), there is a further disadvantage with regard to the other pollutants mentioned above. Using the example of the test series with a swirl number of $S=1.1$, the concentration curves of NH_3 and N_2O can be plotted as a function of the ammonia admixture rate and the equivalence ratio. In general, at the lowest equivalence ratios, the highest N_2O concentrations were found. The NH_3 concentration curves follow this trend with comparatively lower absolute values. However, it can be seen that both N_2O and NH_3 concentrations can be significantly reduced by increasing or decreasing the NH_3 content in the fuel gas. The measured concentrations for pure ammonia combustion in the example mentioned (Figure 8) are a maximum of 30 ppmv N_2O and 5 ppmv NH_3 (NH_3 slip). The low measured values for N_2O when burning pure ammonia cannot be evaluated at present and will need to be put into an overall context in the near future.

Conclusions

The flame stability behaviour of ammonia in combination with natural gas (LNG) and hydrogen was investigated experimentally using both an industrial burner system and a non-premixed laboratory burner.

The measurement campaigns with the industrial burner showed, as expected, the considerable NO_x formation potential in the combustion of methane / hydrogen at already low ammonia admixture rates. In terms of flame stability, the hydrogen-ammonia-blend showed a much more stable combustion compared to the methane-ammonia-mixture, again as expected. In nominal operation, a stable flame could only be maintained for LNG combustion up to $x_{\text{NH}_3} = 0.075$. In contrast, the hydrogen-ammonia-mixture was stable up to $x_{\text{NH}_3} = 0.65$. In the tests with the blend above, a continuous NO_x reduction was also observed for ammonia contents of $x_{\text{NH}_3} \geq 0.4$. The experiments were also repeated at partial load (60 % load) to further investigate the impact of operational settings on flame stability. This change positively influenced the stability, in particular for LNG-ammonia combustion, allowing NH₃ contents up to $x_{\text{NH}_3} = 0.6$ to be used. In the case of the hydrogen-ammonia-mixtures, the partial load operation extended the admixture rate to $x_{\text{NH}_3} = 0.7$.

To investigate higher admixture rates, further experiments were carried out on a specially designed burner test rig. It was possible to stabilise a flame and measure the pollutant emissions at three different swirls for both hydrogen-based and methane-based ammonia blends up to pure ammonia combustion.

The present publication has been deliberately limited to the results of the investigation of flames of methane and ammonia. In the following, the most important results are briefly described:

- Pure ammonia flames could be stably established in a swirl number of $S = 0.5 - 1.1$ at equivalence ratios of $0.83 \leq \phi \leq 1$.

- Low ammonia addition rates increase the NO_x level drastically over those of an e. g. pure methane combustion and show a maximum concentration at approx. $x_{\text{NH}_3} = 0.5$. During the series of measurements carried out, in addition to NO_x, increased N₂O and NH₃ concentrations are also observed in this range.

- An NH₃ admixture of $x_{\text{NH}_3} \geq 0.6$ has a reducing effect on both the resulting NO_x and the N₂O and NH₃ concentrations.

- An increase in the equivalence ratio also led to a reduction in the NO_x concentration in all cases studied.

In summary, ammonia is generally suitable both as an additional fuel gas and for exclusive combustion with air, but require modifications to the burner to achieve stable combustion.

Nevertheless, the results clearly show the need for further research with regard to the further reduction of the resulting emissions, especially nitrogen oxides.

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