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Formation and Control of N2O During Ammonia Combustion: A Summary of What We Know to Date

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Abstract

The development of combustion technologies for carbon free-fuels such as ammonia is expected to enable early realization of a carbon-neutral society. However, among the several challenges of ammonia combustion, the emission of relatively large amount of N_2O, which is a greenhouse gas, at certain combustion conditions undermines the benefits of the carbon-free nature of ammonia. This article summarizes a series of experimental and numerical studies aimed towards the understanding of the formation mechanism and control strategies for N_2O mainly in gas turbines. N_2O is an intermediate species formed along the NO formation pathway. It is rapidly consumed in the flame when the flame temperature is sufficiently high or when there are enough H radicals in the flame. On the contrary, incomplete reduction of N_2O in the flame results in N_2O emissions from combustion devices. A control of N_2O can be achieved through methods that encourage fast and complete combustion of ammonia. We have shown that enhancement of ammonia combustion through co-combustion with other fuels such as methane; a reduction of heat loss from ammonia flames; an elongation of the primary combustion zone of two-stage rich-lean ammonia combustors; and an increase in combustion pressure, strategies which either reduce the reaction time of ammonia in the combustor or increase the residence time of the fuel in the combustion chamber, effectively lead to a decrease in N_2O emissions from ammonia-fueled swirl combustors. This article ends with some perspectives on ammonia combustion in reciprocating engines.

1. Introduction

Ammonia combustion has gained significant attention in the academia and industry in the past couple of years because of the important role ammonia is expected to play in the realization of a carbon-free society. The International Energy Agency (IEA) (IEA, 2021) projects that to achieve a net-zero carbon society by 2050, solar PV and wind would have a combined share of up to 70% of electricity generation. However, given the fluctuations in the supply of these renewables over time, their effective utilization requires the employment of chemical energy storage or carriers in the form of hydrogen or ammonia to cushion the fluctuation. Hydrogen can be produced from the electrolysis of water using power generated from excess renewable energies. On the other hand, ammonia can be produced through Haber-Bosch process using hydrogen and nitrogen obtained through air separation process. Other energy carriers such as methylcyclohexane (MCH), and metal hydrides have been identified for easier transport and storage of hydrogen.

However, in comparison to other hydrogen carriers including liquid hydrogen in compressed and cryogenic forms, ammonia has more promising characteristics such as a higher hydrogen carrying capacity per unit volume, being made of 17.8 % of hydrogen by weight, a higher boiling point than hydrogen which makes it easier to liquify (liquifies at 8.5 bar at room temperature), and an already well-established production and transportation infrastructure. Long distance shipping of hydrogen in the form of ammonia is easier and more economical than its shipping as a pure liquid. On the other hand, ammonia is carbon-free

unlike MCH and is combustible. It is therefore considered that the development of ammonia utilization technologies including in combustion devices is necessary in the carbon-neutral society. The IAE estimates that ammonia and hydrogen will constitute about 2% of electricity production in the net-zero carbon society of 2050 which amounts to about 1360 TWh, while ammonia will have about 46% share of total energy consumption in the maritime sector (IEA, 2021). On the other hand, Japan is encouraging the development of the ammonia fuel industry to facilitate the utilization of ammonia as a fuel in combustion systems, and plans to generate 1% of its total electricity from ammonia/hydrogen by 2030 (METI, 2021).

However, earlier studies of ammonia as a fuel reported challenges associated with ammonia combustion (Pratt, 1967). Ammonia has a low burning velocity that is about five times lower than that of methane, a narrow flammable range, and a high minimum ignition energy. Pratt reported that the low burning velocity inhibits the use of high inlet velocities to achieve effective turbulent mixing in ammonia gas turbine combustors because the fluid residence time in the combustor would be too short for complete combustion of ammonia (Pratt, 1967). However, the need to allow sufficient residence time for ammonia consumption in the combustor by employing low inlet velocities inhibits the mixing of the fuel, air and hot products in the primary combustion zone, leading to a significantly low combustion efficiency. In addition, the low flame speed makes it difficult to stabilize ammonia flames using combustors. On the other hand, the high minimum ignition energy and high ignition temperature present challenges in the ignition of ammonia mixtures in internal combustion engines. Therefore, the light-up of ammonia gas turbines is to date effected by mean of a pilot fuel which is easier to ignite (Kurata et al. 2019).

Recent numerical studies (Somarathne et al., 2017) and experimental investigations (Hayakawa et al. 2017, Okafor et al., 2019a) have however shown that ammonia-air flames can be stabilized over a wide range of conditions by employing swirling flows in combustors. Strong swirling flows generate regions of recirculating fluid motion which circulates hot products and active radicals from downstream to upstream of the combustor. Consequently, mixing and preheating of the fresh unburned mixture is enhanced thereby enhancing the stability of the flame (Syred, 2006). This technology is key in the development of ammonia-fueled gas turbines combustors. Kurata et al. (2017) employed a swirl combustor and successfully generated up to 44 kW for the first time from ammonia-fuelled micro gas turbine with combustion efficiencies of up to 96%. They enhance the combustion of ammonia by preheating the combustion air using a regenerative cycle. On the other hand, co-combustion of ammonia with other fuels with higher burning velocity such as hydrogen, methane, diethyl ether etc. is considered as a means of enhancing the ignition and combustion characteristics of ammonia mixtures in internal combustion engines.

Another important challenge of ammonia combustion application is the emission of large amounts of NOx from the fuel-bound nitrogen atom in ammonia molecule. Fuel NOx production is encouraged by chemical species such as H, O, and OH which promote the conversion of NH_i (i = 0, 1, 2) to NOx mostly via a HNO intermediate (Okafor et al., 2019b). It has been shown that N_2O may constitute a significant part of the NOx emitted from ammonia flames when the flame temperature is relatively low (Okafor et al., 2021a). As an intermediate species during the production of NO, N_2O may be rapidly consumed as it is produced in the flame however low flame temperatures discourage rapid consumption of the molecule. The emission of N_2O, which is a greenhouse gas with a global warming potential of 298 times more than that of CO_2 over a period of 100 years, undermines the carbon-free benefits of ammonia as a fuel.

The control of NOx, including N_2O, from ammonia combustion has been demonstrated using two-stage rich-lean combustion (Okafor 2019a). This NOx control strategy takes advantage of the low NOx production rate from the oxidation of ammonia at fuel-rich conditions. The combustor's primary stage is maintained at a fuel-rich condition to ensure low NOx production. However, this leads to incomplete combustion and the production of large amounts of hydrogen in the flame. Subsequently, air is injected downstream of the combustor to oxidize the unburned ammonia, hydrogen, and other species at an ultra-lean equivalence ratio. Oxidation of ammonia at an ultra-lean equivalence ratio mitigates the formation of NO. However, it may encourage N_2O emissions mainly due to the low oxidation temperature. For instance, studies on the oxidation of mixtures of ammonia and carbon-containing species in staged combustion indicate that HCN and consequently, N_2O production may be important in hydrocarbon–ammonia–air combustion in the ultra-lean secondary stage. Kristensen et al (1996) studied the nitrogen chemistry in the burnout stage of staged combustion containing a mixture of CO, NO, HCN, NH_3 and O_2. They found that HCN oxidation leads to high N_2O production and recorded up to 100 ppmv of N_2O emission at lean burn out conditions. Wargadalam et al. (2000) investigated the homogenous formation of NO and N_2O from the combustion of HCN and NH_3 at 873–1273 K and noted that HCN oxidation promotes NO and N_2O formation. On the other hand, N_2O emission is more significant in liquid ammonia

spray combustion where the evaporation of liquid ammonia droplets encourages heat transfer from the flame thereby discouraging the complete conversion of N_2O in the flame.

It is thus important to understand the formation mechanism and develop control measures for N_2O emission during ammonia combustion to encourage the application of ammonia as a carbon-free fuel. This article discusses a series of studies related to N_2O formation and control conducted by the author at the Institute of Fluid Science of Tohoku University and the National Institute of Advanced Industrial Science and Technology, some of which were conducted in collaboration with IHI Corporation.

2. N_2O in ammonia oxidation chemistry

The chemistry of ammonia oxidation was studied using a detailed reaction mechanism developed by Okafor et al. (2018) which has been optimized for ammonia-air and ammonia-methane-air combustion. Figure 1 shows the pathway for ammonia oxidation in a flame. Ammonia is predominantly consumed through H abstraction reaction with OH, which accounts for more than 70% of ammonia oxidation in the flame. NH_2, NH and subsequently N atom are produced by a progressive abstraction of H atom from the preceding species. In addition to the H abstraction reactions which produce NH_i species with one less H atom in its molecule, the NH_i are oxidized to produce NO directly or through a HNO intermediate. HNO is completely converted to NO through a dissociation reaction and reactions with OH, and H species. These reaction steps which originate from the fuel constitute the major NO production pathways in ammonia flames and are favored by an increase in the concentration of OH, H and O (i.e. O/H) species in the flame. Hence, relatively fuel-lean or near-stoichiometric ammonia flames which are characterized by high concentrations O/H species are associated with high NO production rates. Note also that an increase in flame temperature promotes the production of O/H species through the effect of the temperature-sensitive chain-branching reaction, R1.

$$H + O_2 \leftrightarrow OH + O \qquad\qquad R1$$

Therefore, NO production in ammonia flames increases with an increase in flame temperature however this is not due to thermal NOx production. Initiation of NO production from ammonia flames has its origin in the fuel-bound N atom and occurs when ammonia is oxidized even at relatively low flame temperatures. An increase in flame temperature was found to enhance the NO production pathway relative to its reduction pathway thus leading to an increase in NO concentration with flame temperature.

NO is rapidly converted to N_2O in the flame mainly through reaction with NH radicals in R2, whose reaction rate is mainly controlled by the rate of production of NO in the flame.

$$NH + NO \leftrightarrow N_2O + H \qquad\qquad R2$$

However, numerical analysis of the oxidation of ammonia and methane mixtures in a plug flow reactor shows that at relatively low temperatures below 1300 K, N_2O is produced mainly through the reaction of NH_2 with NO_2 (Okafor et al., 2021b) as represented by R3.

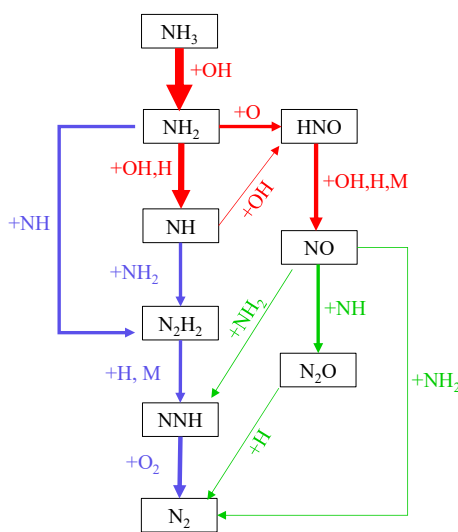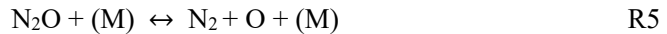$$NH_2 + NO_2 \leftrightarrow N_2O + H_2O \qquad\qquad R3$$

Fig. 1 Ammonia oxidation pathway in a flame. The red, green, and blue pathways represent NO formation, NO reduction and NO-free radical combination pathways, respectively. The reactions shown are only the major reactions in the flame.

On the contrary, N_2O is reduced mainly through reactions R4 and R5;

$$N_2O + H \leftrightarrow N_2 + OH \qquad\qquad R4$$
$$N_2O + (M) \leftrightarrow N_2 + O + (M) \qquad\qquad R5$$

The reaction with H atom has a markedly high temperature sensitivity, hence it is fast in the flame region where temperature is high however the reaction rate drops off for relatively low flame temperatures. On the other hand, the thermal dissociation reaction may continue even at relatively low temperatures but at a very slow rate. It can thus be understood that for conditions with high flame temperatures which are usually accompanied by high H atoms concentration in the flame, N_2O conversion is fast and N_2O emission may be negligible due to complete consumption of the species. On the other hand, at lower temperature the rate of N_2O conversion is slow but continuous via thermal dissociation. In other words, an increase in residence time may ensure complete consumption of N_2O even in flame with relatively low temperatures.

3.   Experimental measurements of N_2O formation

N_2O emissions from the combustion of ammonia-air, and ammonia-methane-air mixtures in swirl combustors, with the ammonia supplied directly to the combustor in gaseous phase or in liquid phase were measured using a Fourier Transform Infrared (FTIR) gas analyzer coupled with O_2 and H_2 analyzers.

3.1   Gaseous ammonia-fueled combustors

Emissions from the combustion of ammonia-air and ammonia-methane-air mixtures were measured using a model swirl combustor of a micro gas turbine which is shown schematically in Fig. 2. The combustor was operated in premixed and non-premixed modes to investigate the effects of mixture uniformity on emissions formation and control.
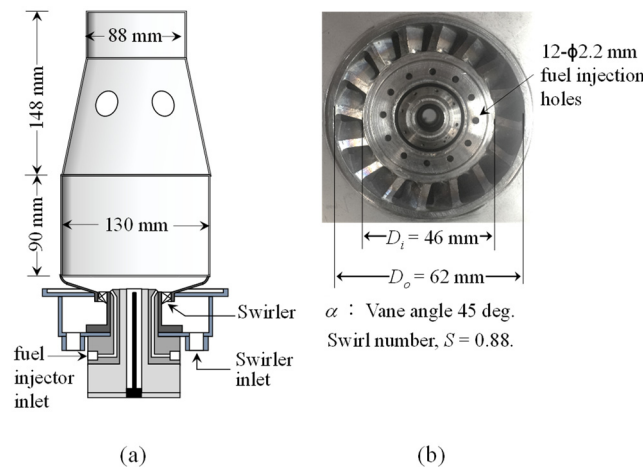

Fig. 2   Schematics of the model swirl combustor of a micro gas turbine. Modified from Okafor et al., 2020.

Figure 3 shows the measured emissions from ammonia-air and ammonia-methane-air flames. The concentration of ammonia in the ammonia-methane binary fuel was expressed in terms of the heat fraction of ammonia, E_{NH3} based on the lower heating value. NO emission was high at fuel-lean conditions. This is due to the relative abundance of O/H species at this condition as explained in the previous section. Observe also that NO emissions from premixed ammonia-methane-air flames were much higher than those from premixed ammonia-air flames owing to higher rate of production of O/H species in the former. At much leaner equivalence ratios below 0.8, a rapid increase in N_2O emissions can be observed which tends to increase with an increase in ammonia fraction in ammonia-methane-air flames. A comparison of N_2O emissions from ammonia-air flames and ammonia-methane-air flames at equivalence ratio of 0.8 shows that pure ammonia combustion has a higher tendency of N_2O emission. A similar rapid increase in N_2O emissions has also been observed in measurements conducted in a stagnation flame burner fueled with ammonia-hydrogen binary fuel as the equivalence ratio became lower than 0.6 (Hayakawa et al., 2022). In

this flame configuration the effects of heat loss from the flame to the stagnation wall was also considered to promote N_2O emission because N_2O emissions increased with a decrease in stagnation wall temperature.
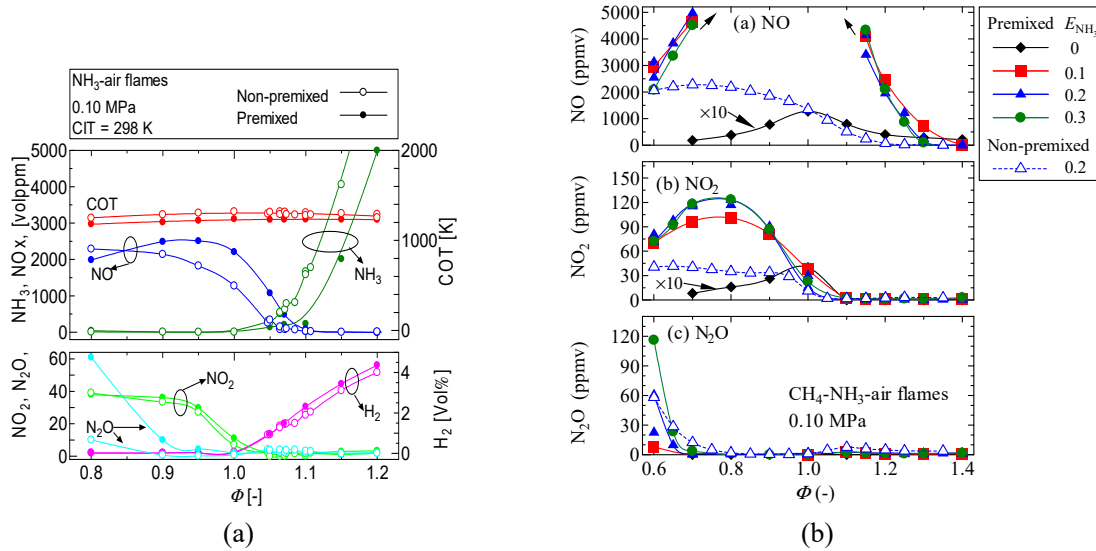

Fig. 3   Measured emissions from; (a) ammonia-air flames (Okafor et al. 2019a), and (b) ammonia-methane-air flames (Okafor et al., 2020). Adopted with permission from Elsevier.

It can be understood from the discussion in Section 2 that at very lean equivalence ratios, H atom concentration as well as the flame temperature is relatively low. Furthermore, the sensitivity of a flame to heat loss may be larger at leaner equivalence ratios. Therefore, rapid consumption of N_2O in the flame region through R4 and R5 is hindered, consequently leading to the measured rapid rise in N_2O emission. Given the low reactivity of ammonia, the effect of heat loss on the flame is expected to be more pronounced in ammonia-air flames than in ammonia-methane-air and ammonia-hydrogen-air flames, hence the higher tendency of N_2O emission from ammonia-air flames.

3.2 Effect of heat loss on ammonia-air flames

The contribution of heat loss from ammonia flames on NOx, including N_2O emissions was investigated using a single stage swirl combustor shown schematically in Fig. 4 a (Okafor et al, 2021a). Three combustor liners were employed which included an insulated steel liner, InSUS; an uninsulated steel liner, UnSUS; and an uninsulated glass liner, UnGLS. The results of the measurement shown in Fig. 4b indicate that N_2O emissions from the uninsulated liners were higher than those from the insulated liner due to wall heat loss. The data plotted here are space-time average values at a point $h_s = 100$ mm above the liner exit. Single point emissions sampling along the radial direction of the combustor liner exit showed that unburned ammonia and N_2O emissions were largest near the combustor wall and negligible near the combustor central axis. This suggests that N_2O consumption was hindered near the combustor walls owing to heat loss. It can be inferred that the usual practice of combustor wall cooling in conventional gas turbines may encourage N_2O emission in ammonia fueled gas turbines, especially at fuel lean conditions where the flame may respond more to heat loss effects.
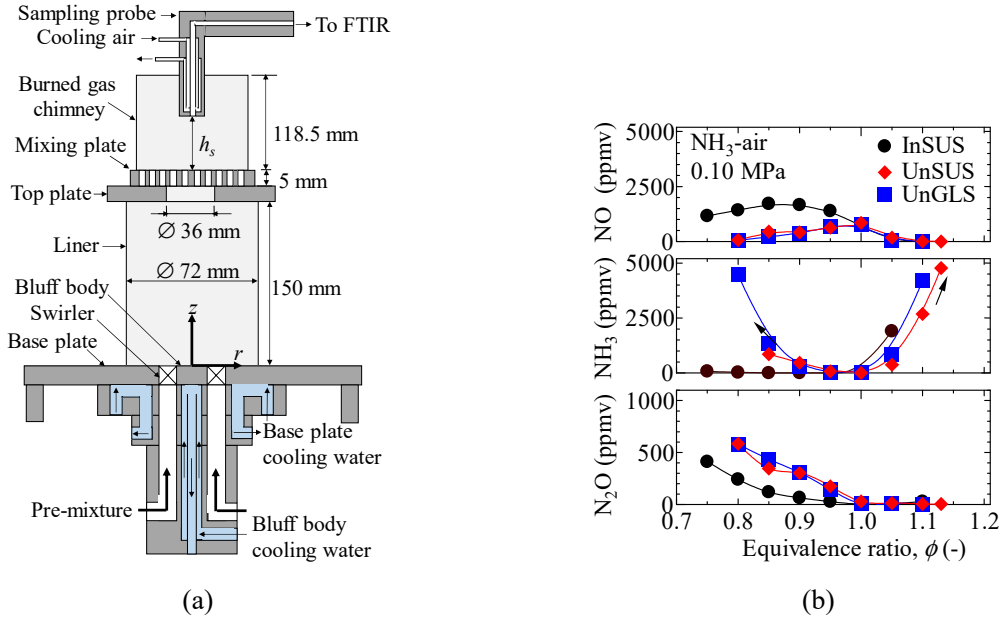
Fig. 4　(a) A schematic of the combustor (b) Comparison of emissions from the combustors. Modified from Okafor et al., 2021a.

3.3　Liquid ammonia spray combustion

The development of liquid ammonia direct spray combustion technology is expected to reduce the capital and running costs of ammonia gas turbines, increase the load per unit volume of fuel supplied and improve the load-following capabilities of the gas turbines. However, owing to the high latent heat of vaporization of liquid ammonia which is about 7% of the lower heating value, a significant amount of heat transfer from the flame to the vaporizing liquid droplets occurs during liquid ammonia spray combustion, hindering flame stabilization and encouraging N_2O emissions. Poor flame stability was a reason previous research efforts to investigate liquid ammonia spray flame in gas turbine combustor were unsuccessful.

However, by injecting liquid ammonia in a strongly swirling flow of preheated air, we achieved the first ever successful stabilization of pure liquid ammonia in a gas turbine combustor at elevated pressures. Preheating of the air was necessary for flame stabilization as it compensated for the latent heat of vaporization. In addition, further improvement of flame stability was achieved by co-firing the spray flame with methane. Figure 5a shows the effects of air preheat temperature on measured emissions from the combustor. An increase in preheat temperature resulted in a decrease in N_2O and other emissions from the combustor. On the other hand, Fig. 5b compares the emissions of N_2O from pure liquid ammonia spray flame and liquid ammonia co-fired with 50% methane by lower heating value. Observe that N_2O emissions from the pure liquid ammonia spray was markedly higher than that from the co-combustion of the spray and methane. In addition, the emission of N_2O increased with an increase in input thermal power for both fuels. It can be understood that the higher rate of heat release from the methane chemistry may reduce the response to the flame to heat transfer and hence enable the consumption of N_2O in the flame. On the other hand, an increase in input thermal power encourages an increase in liquid mass loading which may promote group droplet combustion in the flame. Consequently, longer droplet lifetime and local decrease in flame temperature are promoted thereby encouraging N_2O emissions.
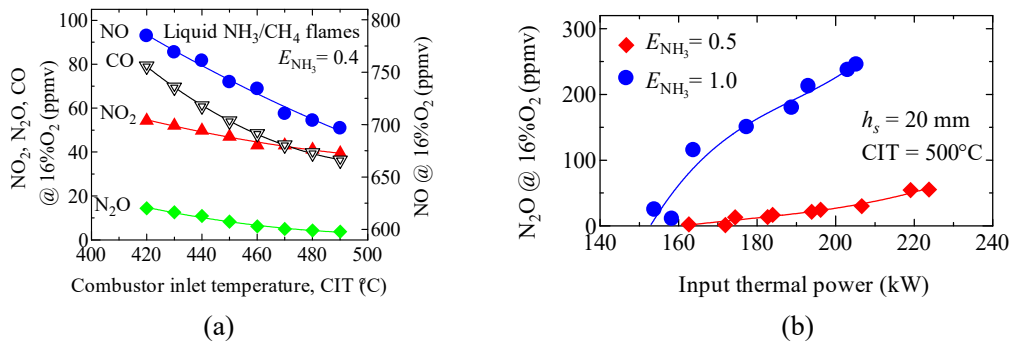
Fig. 5   (a) Effects of air preheat temperature on emissions from a gas turbine combustor co-firing liquid ammonia spray and methane (b) A comparison of N_2O emissions from pure liquid ammonia spray flame and liquid ammonia spray co-fired with methane. Modified from Okafor et al., 2021c.
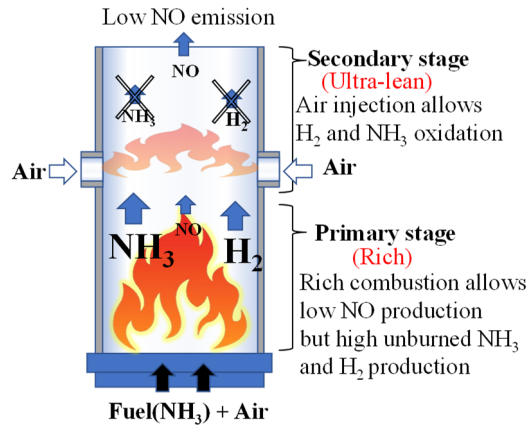

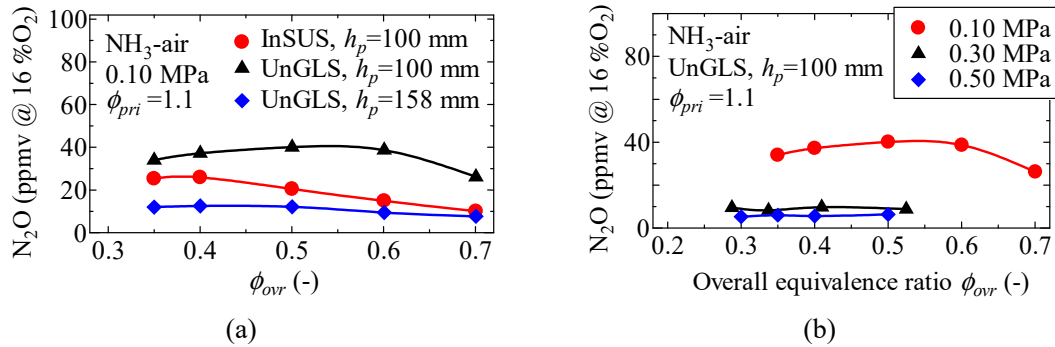Fig. 6   A schematic of two-stage rich-lean combustion concept

Fig. 7   A comparison of the effects of (a) wall heat loss and height of the primary stage, and (b) combustion pressure on N_2O emissions at primary equivalence ratio, ϕ_{pri} = 1.1. Modified from Okafor et al. 2021.

4. N_2O emissions control strategies

The control of NOx emission from gaseous ammonia combustion has been demonstrated using two-stage rich-lean combustors numerically by (Somarathne et al., 2017) and experimentally by (Okafor et al., 2019a, 2020), and (Kurata et al., 2019). Figure 6 shows schematics of the concept of two-stage rich-lean combustion. This strategy has also been applied to liquid ammonia spray combustion (Okafor et al., 2021c).

We investigated the effects of combustor wall heat loss, the height of the primary stage, h_p and ambient pressure on N_2O control from two-stage rich-lean combustors. Emissions from gaseous ammonia-air flames were measured in a laboratory scale experiment using three model swirl combustor types viz-a-viz an insulated combustor with h_p =100 mm; an uninsulated combustor with h_p =100 mm; and an uninsulated combustor with h_p = 158 mm. The measurements were conducted for pressures up to 0.5 MPa (Okafor et al. 2021a). Figure 7a shows the measured N_2O emissions from the combustors at 0.10 MPa. The insulted combustor of h_p = 100 mm had lower N_2O emissions than the uninsulated one with the same h_p. This demonstrates that a control of N_2O emission can be achieved by reducing heat loss from ammonia fueled combustors. Furthermore, the data plotted in Fig. 4b also indicates the importance of a reduction in heat loss for the control of N_2O emissions from combustors. Similarly, studies conducted in a 50 kW type gas turbine combustor test rig fueled with liquid ammonia spray showed that a two-stage combustor without wall film cooling resulted in lower N_2O emissions than a similar combustor with wall film (Okafor et al., 2021c).

On the other hand, Fig. 7a show that the uninsulated liner of h_p = 158 mm had lower N_2O emission than the uninsulated liner of h_p = 100 mm. An increase in fluid residence time in the combustor even in the presence of heat loss is expected to encourage an increase in the dissociation of N_2O through R5. On the other hand, the secondary air which dilutes the flame in the secondary stage encourages a decrease in the combustion temperature. This may promote N_2O production in the secondary stage especially when there is a sufficiently large amount of unburned fuel species in the secondary stage. However, the more the height of the primary stage, the higher the combustion efficiency in that stage and thus the lower the amount of unburned fuel and NH_i species that encounter the injected cold secondary air. Hence, an elongation of the primary combustion zone is an effective control strategy for N_2O even in the presence of heat loss from the flame. Figure 7a shows that an increase in h_p was a more effective N_2O control strategy than insulation of the combustor wall.

Figure 7b shows the effects of pressure on the emissions of N_2O from the uninsulated liner of h_p = 100 mm. N_2O emission decreased with an increase in pressure. Note that in these measurements, the combustor inlet velocity was kept constant, therefore the mass flow rates increased with pressure for each equivalence ratio. Because the mass burning flux increased with pressure due to increased density and turbulence, the associated increase in heat release rate may encourage a decrease the effect of heat loss on the flame and consequently result in lower emissions of N_2O. On the other hand, an increase in combustion efficiency with pressure may reduce the contribution of the injected secondary air to N_2O emission as explained above.

5. Perspectives on N_2O emission formation and control in reciprocating engines

Efforts are on-going to develop technologies for reciprocating engines fueled with ammonia, particularly for maritime transport. Among the major challenges of this technology include the emissions of high levels of unburned ammonia and N_2O trapped in crevices in the combustion chamber. Because ammonia has a relatively large quenching distance, it is expected that the flame will be quenched relatively farther from the chamber wall, leaving regions of unburned fuel close to the wall. Such quenching of ammonia flame in the engine cylinder would inhibit the consumption of intermediate species such as N_2O. Therefore, it suffices to expect a high concentration of N_2O emitted along with unburned ammonia from the engines.

On the other hand, ammonia-air mixtures have Lewis numbers that are comparable to those of methane-air mixtures. However, due to the larger flame thickness of ammonia, ammonia flames have a higher Markstein number and a lower extinction stretch rate. For the same turbulence intensity, ammonia flames also possess a higher turbulence Karlovitz number than methane flames. In other words, ammonia flames have more tendency to be quenched in the chamber because of stretch. Therefore, local flame quenching may contribute to unburned fuel and N_2O emissions from ammonia-fueled reciprocating engines.

It is therefore considered that co-combustion of ammonia and other fuels with higher flame speeds is necessary in reciprocating engines for the purpose of improving the combustion efficiency and controlling N_2O emissions. Reciprocating engines may benefit from in-situ partial cracking of ammonia along the fuel supply line which yields to a dual fuel mixture of ammonia and hydrogen. However, even though an enhancement of ammonia combustion may reduce N_2O and unburned fuel emissions, it may encourage the emission of NO due to an increase in flame temperature and the rate of production of O/H radicals.

6. Summary and conclusion

The emission of N$_2$O from ammonia combustion systems undermines the benefits of the carbon-free characteristics of ammonia as a fuel and may discourage the application of ammonia for direct reduction of greenhouse gases emission. The chemistry of N$_2$O formation in ammonia flames has been explored in this article. N$_2$O which is an intermediate species in the NO production pathway can be consumed as fast as it is produced when the flame temperature or H atom concentration in the flame is sufficiently high. Furthermore, experimental studies have shown that N$_2$O emissions are encouraged by conditions that enable slow and incomplete combustion of ammonia such as heat loss from the flame. In other words, a control of N$_2$O emissions from ammonia combustion systems can be achieved by promoting fast and complete combustion of ammonia through a reduction of heat loss from the flame, co-combustion of ammonia with other fuels with higher rate of heat release and high-pressure combustion. These have been demonstrated in the experimental studies summarized in this article.

Acknowledgements

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