INVESTIGATION OF NO EMISSION CHARACTERISTIC OF AMMONIA-HYDROGEN FLAME IN A TWO-STAGE MODEL COMBUSTOR

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The laminar burning velocity and NO formation process of ammoniahydrogen combustion within a two-stage combustion chamber were investigated numerically in the present study. A Chemical Reactor Network (CRN) method involving perfectly stirred reactor (PSR), plug flow reactor (PFR), and partially stirred reactor (PaSR) configurations with the 24-species Xiao mechanism was implemented to simulate the premixed ammonia-hydrogen-air combustion process. The effects of inlet temperature and pressure conditions on the laminar burning velocity were investigated. Results proved that elevated pressure condition decreased primary flame thickness leading to lower laminar burning velocity while inlet temperature increased flame temperature which in turn increased the laminar burning velocity. Investigation of the effect of humidification on the laminar burning velocity showed that humidification can counteract the effect of high inlet temperature. NO emission studies indicated a twofold impact of pressure on NO formation processes: (1) preventing NO formation in the primary combustion zone; (2) promoting thermal NO formation in the lean combustion zone. The minimum amounts of NO emission were obtained at total equivalence ratios of 0.4. Humidification prevented the NO formation in the lean combustion through the competitive effect of H_2O on O, whilst temperature effect was comparatively small. Humidity and pressure were optimized in the two-stage configuration to achieve both low emission and high efficiency.

Key Words: Ammonia-hydrogen combustion, Two-stage combustion, Chemical Reactor Network, Laminar burning velocity, NO emission

1. Introduction

Ammonia as a carbon-free fuel has been recently used as a promising component in blends fuels by the International Energy Agency [1, 2]. Ammonia has high hydrogen content (17% mass) [3] and its combustion products only consist of nitrogen and water in completely burning condition.

Furthermore, ammonia is the second most bulk manufactured chemical, and it has been mass produced throughout the world for more than a century [4]. Therefore there are already numerous infrastructures and much experience in regard to its handling. However, some remarkable combustion challenges are associated with the utilization of NH₃, mainly due to its low burning velocity and high ignition energy, in addition to the large amount of NO_X emission that its combustion can produce [5, 6].

Several experimental and numerical studies have been conducted on ammonia fueled gas turbines systems with pure ammonia [6, 7], methane-ammonia [8-12], coke oven gas (COG)-ammonia [13], and ammonia-hydrogen [14], showing that optimum operation for low NO_x emission can be achieved with rich combustion conditions at the primary combustion zone and ammonia has an effective role in the reduction of NO [15, 16]. Supplementary fuels such as H₂ have been shown to improve and stabilize operational performance [17, 18]. Valera-Medina et al. [19] conducted several experiments within a gas turbine environment for NH₃-H₂ blends demonstrating that 50%NH₃/50%H₂ blend fuel can effectively improve the laminar burning velocity. However, this blend narrows flame stability limits due to the high diffusivity of hydrogen. Further tests conducted by the group showed that less hydrogen (30% molar fraction) can provide stable combustion profiles [20]. Nevertheless, one impediment of using blend fuels of ammonia and active reactants in practical equipment is linked with the high NO_X emission they can produce. Although the fuel rich condition in ammonia containing fueled gas turbine system is confirmed to be advantageous for low NO_X emission, it is accompanied with lower combustion efficiencies (represented by some unburned ammonia). Therefore an efficient combustion system that ensures NO_x emissions reduction with combustion efficiency improvement is essential.

Two-stage combustion has attracted attention of researchers as an effective system recently [21-24], which can permit the reduction of NO_X emissions and utilize the fuel more completely. Two-stage combustion systems fueled by NH₃ and NH₃-H₂ were investigated experimentally in a gas turbine power generation system, and the results show that unburnt NH3 released from the fuel rich region reduced NO emission from the fuel lean region in the secondary combustion zone through the selective non-catalytic reduction (SNCR) mechanisms. This effect denoted increasing NO emission firstly, which then decreased with the increase of NH₃ content [21]. Additionally, the effects of primary equivalence ratio, total equivalence ratio, inlet temperature and combustion chamber pressure on NO emission in two-stage combustion systems with ammonia-containing fuel were also studied [22, 23]. In further implementations, on the other hand, the injection of steam into gaseous ammonia combustors has been reported to be effective in the control of NOx emissions. Pugh et al. [24] reported that steam injection inhibited NO production from gaseous ammonia-hydrogen-air flames due to the combined influences of a reduced temperature and enhanced O atom consumption through its reaction with H₂O molecules to inhibit N₂+O<=>NO+N. The above-mentioned investigations demonstrate that the two-stage combustion technology can be a promising method for ammonia-containing combustion with low emissions and high efficiency.

The focus of this study is on the laminar flame stabilization and NOx emission of the premixed ammonia-hydrogen-air combustion in a two-stage combustor. This work intends to develop a numerical scheme to simulate the flame propagation and NOx formation processes in a combustion chamber containing two stages through a systematic study over a wide range of equivalence ratio, humidified fraction and inlet temperature at elevated pressure condition.

2. Methods and materials

A fuel composition with an NH_3 - H_2 volume ratio of 70%/30% was specified for this paper [20]. The inlet temperature was set in the range of 400 K to 600 K, and the pressure condition was specified at 1.0, 2.0, 3.0 MPa, respectively. The primary equivalence ratio and total equivalence ratio are varied from 1.1 to 1.5, and from 0.4 to 1, respectively. Water steam was introduced only into the primary combustion zone with primary airflow at four distinct humidified volume fractions: 0 %, 4 %, 8 %, 12 %. Furthermore primary and total equivalence ratio were calculated using the following equation:

$$\Phi_{\text{primary}} = \frac{(\text{fuel / air})_{\text{primary}}}{(\text{fuel / air})_{\text{primary stoic}}}$$
(1)

and

$$\Phi_{\text{total}} = \frac{(\text{fuel / air})_{\text{primary+secondary}}}{(\text{fuel / air})_{\text{primary+secondary stoic}}}$$
(2)

from

$$0.3H_2 + 0.7NH_3 + 0.675O_2 + 2.54N_2 \rightarrow 1.35H_2O + 2.89N_2$$
 (3)

All examined conditions are summarized in Table 1.

In order to determine the primary laminar burning velocity as a critical parameter for stable combustion system development, this parameter was evaluated at different T_i value using Cantera [25]. The analysis was carried out using a reduced model of Xiao mechanism containing 24 species and 91 steps [26]. Results of Xiao mechanism are consistent with experimental data in prediction of laminar burning velocity and it can be used at elevated pressure conditions [27]. In addition, Mao *et al.* [27] showed an approximate NO concentration value at the same condition. Hence the Xiao mechanism is used for the analysis of NO emission for NH₃-H₂ fuel blend combustion in the present work.

Table 1 Examined conditions.

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Composition (Volume Fraction)	$70\% \text{NH}_3 / 30\% \text{H}_2$
Fuel Mass Flow Rate (g/s)	1.388
$oldsymbol{arPhi}_{ ext{primary}}$	1.10, 1.20, 1.30, 1.40,1.50
$oldsymbol{arPhi}_{ ext{total}}$	0.4, 0.5, 0.6, 0.7, 0.8, 0.9, 1.0
Humidified Volume Fraction	0%, 4%, 8%, 12%
Pressure (MPa)	1.0, 2.0, 3.0
2nd Air Temperature (K)	300

A chemical kinetic modelling tool, CHEMKIN-PRO [28] was utilized to simulate the NO emission of the two-stage combustion system. The simulations were conducted using a chemical reactor network (CRN) method formed by a hybrid perfectly stirred reactor (PSR), plug flow reactor (PFR), and partially stirred reactor (PaSR) configuration [29-31]. A schematic of the network employed in the current study, including its four clusters, is presented in fig.1. Two inlets were used to provide ammonia-hydrogen fuel mixture, and air/water steam mixture, with three PSR to model premixing, combustion, and central recirculation zone. The post flame zone was modeled by a PFR with one dimensional length of 5 cm. The quenching/mixing zone designed for mixing the products from the rich combustion zone and secondary air, was modeled by a PaSR. The second PFR with one dimensional length of 5 cm was used to simulate the lean combustion [31].

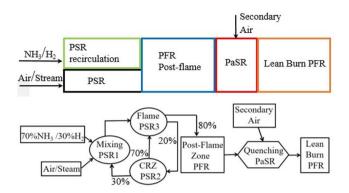


Figure 1. 1D Chemical Reactor Network schematic.

3. Results and discussion

3.1 Laminar burning velocity

Laminar burning velocity is one of the most important global characteristics of fuel combustion, which can be used to characterize many premixed flame phenomena, such as chemical reaction characteristic time, flame stability, flame height. In the two-stage combustion system, the main flame zone is in the state of rich combustion, while the laminar combustion velocity of NH_3 containing fuel decreases with the increase of equivalent ratio on the rich combustion side, so that the flame of NH_3 -containing fuel tends to be unstable and blowout occurs. Therefore the laminar burning speed must reach a reasonable range to keep the flame stable.

Figure 2 shows that the primary laminar burning velocity decreases with the increase of primary equivalent ratio, under different inlet temperatures and pressures. On the other hand, the laminar burning velocity increases monotonically with the decreases of pressure and the increase of inlet temperature, as reported in [27, 32]. This can be explained by the concentration of OH radicals [26] which are mainly produced near the flame front and influenced by the thickness and temperature of the flame.

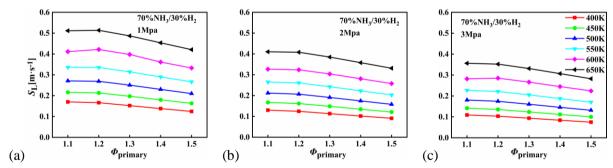


Figure 1. Primary laminar burning velocity against primary equivalence ratio with different inlet temperature at P = 1.0, 2.0, 3.0 MPa.

Figure 3 shows that the OH mole fraction increases dramatically with the increase of inlet temperature, and decreases relatively gently with the increase of pressure. The inlet temperature plays a more important role in primary laminar burning velocity than pressure. Increasing inlet temperature and pressure can both increase flame temperature and decrease the_flame thickness. However, increasing the pressure increases the flame temperature very slightly, but significantly decreases the flame thickness, which demonstrates how pressure reduces primary laminar burning velocity mainly by thinning the flame front, and then decreasing the OH mole fraction. Comparing the effect of increasing inlet temperature on the reduction of flame thickness, the influence of inlet temperature on flame temperature dominates the OH behavior. Hence, adequate reactivity can be ensured by increasing the inlet temperature to effectively mitigate the decrease in laminar combustion rate caused

by increased pressure.

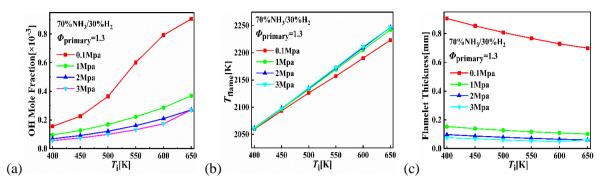


Figure 2. OH mole fraction, primary flame temperature and primary flame thickness against inlet temperature with different pressure at $\phi_{\text{primary}} = 1.3$ in primary combustion zone.

Figure 4 compares the variations of primary laminar burning velocity with inlet temperature for different humidified fractions and pressures. The primary laminar burning velocity of NH_3 - H_2 increases with the increase of inlet temperature under humidified condition. Water steam decreases the flame temperature and increases the flame thickness. The reduction of flame temperature contributes mostly to the reduction of OH mole fraction. The decrease of laminar burning velocity caused by water steam can be compensated by increasing inlet temperature.

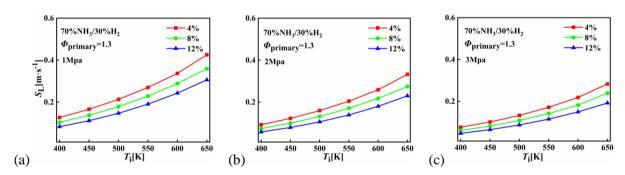


Figure 3. Primary laminar burning velocity against inlet temperature with different humidified fraction at P = 1.0, 2.0, 3.0 MPa.

3.2 NO Emission

Additionally to the above mentioned laminar burning velocity, NO emission is another important parameter for the usage of ammonia-containing fuels due to the nitrogen atom in the ammonia molecule. Figure 5 shows the variation of NO mole fraction with total equivalence ratio for different primary equivalence ratios at elevated pressures of P=1.0, 2.0, 3.0MPa. NO emission increases first and then decreases with the increase of total equivalent ratio. This is attributed to NH_i in the reduction of NO, alongside the role of air in thermal NO production. In addition, with the increase of pressure, the overall NO emission increased. It can be observed as well that the peak location of NO emission with larger primary equivalent ratio moved to the lower total equivalence ratio side.

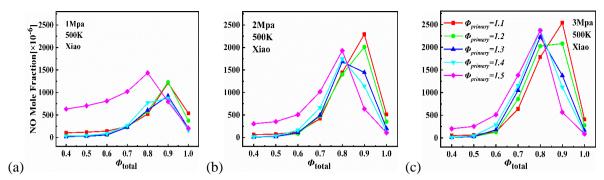


Figure 5. NO emission against total equivalence ratio with different primary equivalence ratio at P = 1.0, 2.0, 3.0 Mpa.

To conduct the kinetic analysis further and explore the reason of this phenomenon, the Xiao mechanism was used to study the main reaction paths of NO formation in different regions under different pressures. It is known that two-stage combustion can reduce NO emissions and improve combustion efficiency in an overall lean condition, but may produce more NO emission in comparison with single-stage rich combustion due to the NO formation in both combustion zones [24].

On the one hand, elevated pressure inhibits NO emission in the primary combustion zone, as summarized in Table 2. Figure 6(a) shows that the reaction NO₂+H<=>NO+OH, which controls the production of NO, is offset by the nearly equivalent NO consumption reaction NO+HO₂<=>NO₂+OH at the flame zone pressure up to 3 Mpa, ultimately leading to a marginal reduction in NO fraction in the primary flame zone. However, NO+H(+M)<=>HNO(+M) plays an important role in the formation of NO when the pressure is 1 Mpa and 2 Mpa, and the formation rate increases with the increase of pressure. This phenomenon can be explained by the contraction of the reaction zone and the reduction of the OH radical pool. The OH radicals act as the direct reactant to form NO, or react first to form HNO and then to produce NO, thus NO emission is very sensitive to OH radicals [30]. In addition, although the total NO net production rate in the primary flame zone increases with pressure, the decrease of NO emission means that the reaction zone is thinned. NO reduction takes place in the post flame zone, and the reduction rate decreases when the pressure increases, as presented in fig. 6(b).

On the other hand, NO formation is mainly through thermal NO formation mechanisms in the lean combustion zone, fig. 6(c)-6(d). And the comparison of absolute rate of NO production shows that NO formation is strengthened with the increase of pressure, causing lean combustion zone produce more NO for higher pressure. The competition of the opposed effects of pressure in the two combustion zone systems determines the ultimate NO emission. When total equivalence ratio is lower than 0.6, the excess air cools down lean combustion zone effectively to suppress the thermal NO formation.

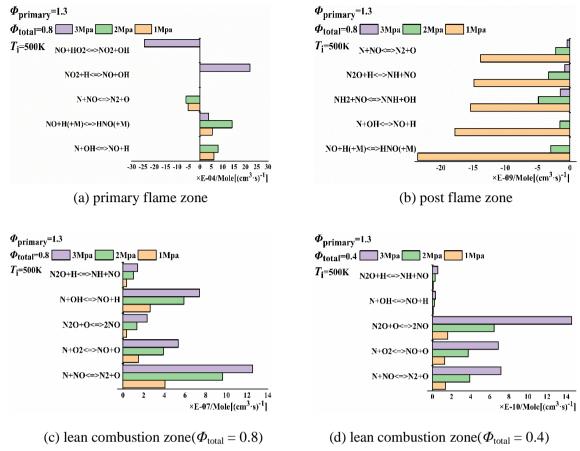


Figure 6. Absolute rate of NO production for elevated pressure.

Compared to the single-stage rich combustion, two-stage combustion can achieve a slightly lower NO concentration at $\Phi_{\text{total}} = 0.4$ for the same rich condition. But a moderate air in the lean combustion zone provides an appropriate temperature and oxidizer to promote NO formation for total equivalence ratios between 0.7 and 1.0. Thus NO formation in the lean combustion zone dominates the ultimate NO emission with NO mole fraction approaching a peak around $\Phi_{\text{total}} = 0.8$. To sum up, elevated pressure reduces the NO emission in the primary combustion zone effectively, and the total equivalence ratio needs to be lower than 0.6 to suppress the thermal NO formation in the lean combustion zone.

Table 2. NO mole fraction from CHEMKIN-PRO applied to the two stage combustion system.

Pressure	Primary flame	Post flame	Quenching zone	Lean combustion
	zone	zone		zone
1.0 Mpa	1.25E-03	2.45E-04	2.09E-04	6.61E-04
2.0 Mpa	8.19E-04	6.03E-05	5.13E-05	1.24E-03
3.0 Mpa	6.19E-04	3.68E-05	3.14E-05	1.74E-03

To illustrate the importance of the reaction paths close to the flame zone, a sensitivity analysis was performed. As shown in fig. 7, it is clear that the combination of NO_2 species with other radicals tends to play a positive role, while most of the NO reactions with HO_2 have a negative impact on the production of NO. Interestingly, the reaction NO+OH(+M)=HONO(+M) can also be another path towards the reduction of nitrogen oxides in these flames.

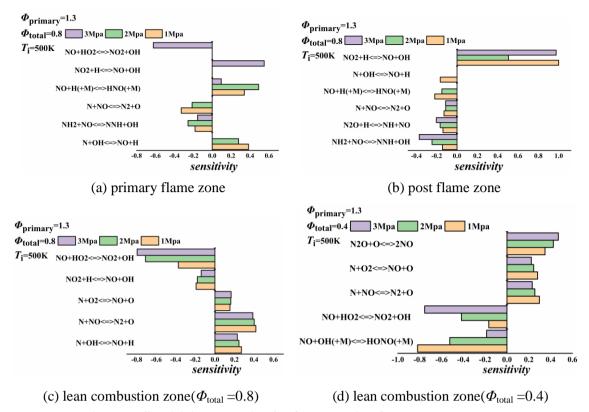


Figure 7. Sensitivity analysis of NO production for elevated pressure

NO emission with different amounts of water steam injection at P = 3 MPa and $T_i = 500$ K was also analyzed, as shown in fig. 8. Similar to the condition without humidification, total equivalence ratio plays an important role in NO concentration at humidified conditions. But it can be observed that addition of water steam decreases the NO concentration for total equivalence ratios between 0.9 and 1.0. The effect of water steam addition on the NO emission can be divided into two part: the primary combustion zone and the lean combustion zone.

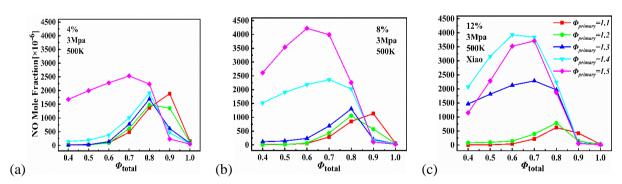


Figure 8. NO emission against total equivalence ratio with different primary equivalence ratio at 4%, 8%, 12% humidified volume fraction at P = 3.0 MPa and $T_i = 500$ K.

Figure 9(a)-9(b) shows a comparison between the absolute rates of NO fractions produced from prominent reactions in the flame zone and in the post-flame zone, with changes in humidification fraction. The results reveal that the most influential promoting reaction is NO+H(+M) \ll +NO(+M) and the most inhibiting reaction is NH₂+NO \ll +NNH+OH. They have the largest impacts below 3 MPa, and dominate the NO production and consumption as humidification fraction rises in the flame

zone. This phenomenon may be a consequence of the increased concentration of the OH free radical pool, whereas enhanced NO consumption, in the flame zone for 4% water fraction, results from similar mechanisms to those shown for an increase in pressure at 3 Mpa (the reactions discussed in Fig. 6).

Furthermore, the secondary air lowers the temperature in the lean combustion zone, suppressing the thermal NO formation. Meanwhile, the content of N and O increases, which leads to the oxidation of NO. However, at this stage, the H₂O entering continuously competes with N and consumes O through the reaction $O + H_2O \ll OH + OH$. The $NH_3 + OH \ll NH_2 + H_2O$ reaction generates NH_1 through the chain carrying mechanism with OH, and the reduction effect suppresses the generation of NO. Thus, the competitive effect of H₂O and N atoms on O atoms determines the formation of NO. As shown for $\Phi_{\text{total}} = 0.9$ in fig. 9(c), O atom is continuously consumed by H₂O, and the reducing ability of H atom and NH_i generated by chain reaction is enhanced, while the overall NO decreases. Conversely, as shown for $\Phi_{\text{total}} = 0.4$ in fig. 9(d), the reduction of NO due to the consumption of H₂O is less than the production of NO due to the oxidation of N with the increase of O atom in the secondary air, consequently the total number of NO increases. However, under this condition, temperature has little effect on the production of thermal NO. In addition, a noticeable difference between the two conditions is that: $NO + H(+M) \le HNO + (+M)$ is the most inhibiting reaction in Φ_{total} =0.9, whilst NO + OH (+M) <=> HONO (+M) is the most inhibiting reaction in Φ_{total} =0.4. However, both of these element reactions are positively correlated with NO emission. Therefore HNO and HONO elements both play an important role in NO emission.

According to the above analysis, due to the different mechanisms controlling NO formation, secondary air and water effects can be combined to provide a greater overall reduction of NO while leading to a reduction of unburned NH_3 for higher combustion efficiency. Therefore under humidification conditions, a Φ_{total} can be used to reduce unburned fuel fraction without NO emission penalty, which is particularly important for combustion of gas turbine under real conditions.

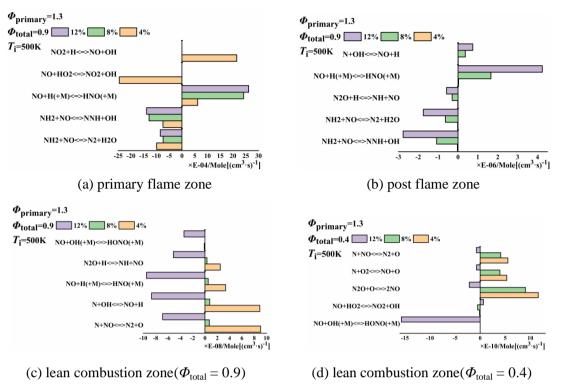


Figure 9. Absolute rate of NO production at different humidified fraction with Xiao mechanism for P = 3.0 MPa and $T_i = 500$ K.

4. Conclusion

A numerical analysis using a chemical reactor network method was performed in order to inspect the feasible laminar burning velocity and potential to minimize the NO emission, at elevated pressure conditions with humidified primary air, for NH_3 - H_2 fuel blends in a two-stage combustion system. Results proved that elevated pressure thinned the flame thickness causing a decrease in the OH radical concentration, thus lowering the primary laminar burning velocity. The investigation of the effect of inlet temperature showed that higher inlet temperature increased the primary laminar burning velocity by increasing flame temperature. Humidification can counteract the effect of increasing inlet temperature on laminar flame velocity. An increase in pressure results in lower exhaust NO concentrations in the primary combustion zone by thinning the flame front. On the contrary, the temperature effect obviously promoted the formation of thermal NO. Ultimately, thermal NO formation dominated the NO formation, and $\Phi_{\text{total}} = 0.4$ was an optimal overall condition to get a low NO emission. Reactant humidification reduces NO concentrations in the lean combustion zone through the competitive effect of H_2O on O. Therefore the combination of humidification and secondary air flow can be used to improve the emissions performance of an NH_3 - H_2 at elevated pressure conditions.

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