

EVALUATION OF MINIMUM NO_x EMISSION FROM AMMONIA COMBUSTION

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ABSTRACT

Ammonia (NH₃) is being explored as a hydrogen carrier with no carbon emissions. However, the fuel-bound nitrogen when burning ammonia comes with a potentially significant NO_x emissions penalty. This work aims to establish the theoretical minimum NO_x emissions for an axially staged ammonia combustor, to determine how much NO_x levels can be reduced via further technology development. The optimum residence time and stoichiometry of each zone is determined in the fast mixing limit with a constraint on unburned H₂ at the exit. These results show minimum NO_x levels are in the 195-400 ppm range at 1 bar but drop to levels of ~25 ppm at 20 bar. Results suggest that NO_x values drop with increased combustor residence time, increased pressure, and increased combustor exit temperature. These NO_x emissions are dominated by NO_x production in the primary stage which relaxes to equilibrium levels quite slowly, suggesting that promising routes for research are kinetic means to accelerate primary zone NO_x relaxation rate to equilibrium. They also show that combustors specifically designed for ammonia will have residence times that are one to two orders of magnitude larger than current natural gas systems. By doing so, NO_x levels below 10 ppm may be achievable.

Keywords: ammonia combustion, NO_x emission, reactor network modeling

NOMENCLATURE

CRN	Chemical Reaction Network
H ₂	Hydrogen
NH ₃	Ammonia
N ₂	Nitrogen
NO _x	Nitrogen Oxides
O ₂	Oxygen
RQL	Rich Quench Lean
τ	residence time
Φ	equivalence ratio

INTRODUCTION

Ammonia (NH₃) is being actively investigated as a carbon-free alternative fuel. However, the fuel-bound nitrogen in NH₃ causes a significant increase in NO_x emissions during ammonia combustion (Elbaz et al., 2022; Kobayashi et al., 2019; Valera-Medina et al., 2018). Ammonia has widespread applications related to fertilization and refrigeration, which have yielded an infrastructure for the production and transportation of ammonia (Kobayashi et al., 2019). However, ammonia has challenging combustion characteristics including a limited flammability range, lower flame temperature, and slower laminar flame speeds as opposed to traditional hydrocarbon fuels. Furthermore, NO_x emissions from the fuel-bound nitrogen pose a significant challenge to the viability of ammonia fuels, as these emissions may offset the benefits of being a carbon-free fuel. Fundamental studies of NO_x formation in ammonia combustion (Mashruk et al., 2022a; Mashruk et al., 2022b; Pugh et al., 2021; Mashruk et al., 2022c; Viguera-Zúñiga et al., 2021), and minimal emissions ammonia combustor configurations (Mohammadpour et al., 2022; Rocha et al., 2021; Pugh et al., 2021; Khateeb et al., 2021; Li et al., 2022; Okafor et al., 2021) are integral research contributions to evaluate ammonia's potential as a carbon-free fuel.

As opposed to conventional combustors designed for hydrocarbon fuels, it is still unclear what strategy and structure to use for ammonia-based combustor design, therefore, a combustor suitable for ammonia combustion still does not exist. Different strategies have been employed, including swirl stabilization to hold the low flame speed mixture (Mashruk et al., 2022a; Mashruk et al., 2022b; Pugh et al., 2021; Mashruk et al., 2022c; Viguera-Zúñiga et al., 2021), MILD and flameless combustion (Mohammadpour et al., 2022; Rocha et al., 2021), ammonia fuel blends (Mashruk et al., 2022a; Mashruk et al., 2022b; Pugh et al., 2021; Mashruk et al., 2022c; Viguera-Zúñiga et al., 2021;

Mohammadpour et al., 2022; Rocha et al., 2021), elevated pressures (Rocha et al., 2021; Pugh et al., 2021; Khateeb et al., 2021), and staged combustion or Rich-Quench-Lean (RQL) (Rocha et al., 2021; Li et al., 2022; Okafor et al., 2021; Okafor et al., 2019).

Staged combustion systems are commonly used today for enhancing turndown and reducing NO_x . In natural gas fueled systems, a secondary fuel stage (e.g., an “axial fuel stage”) is used to minimize NO_x emissions at elevated combustor exit temperatures. In these systems, the majority of the fuel is burned in a lean, premixed mode in the primary zone at a low temperature to avoid significant NO_x production. Then, the remaining fuel is added in a second lean stage, where quick mixing and combustion is desired in order to minimize residence times at high temperature, while still consuming all the fuel (Rocha et al., 2021). Rich primary zone, followed by a lean secondary zone (RQL) systems are used for minimizing NO_x for systems with high turndown (McKinney et al., 2013) and/or with systems with fuel-bound nitrogen. In the latter case, the abundant NH_2 radical produced in rich flame reacts with NO to reduce NO_x emission. Therefore, staged combustion with a rich flame zone then lean flame zone has been investigated to reduce NO_x levels from ammonia combustion (Li et al., 2022; Okafor et al., 2021; Okafor et al., 2019; Wu et al., 2020; Mashruk et al., 2021; Guteša Božo et al., 2021). Such an RQL configuration leads to substantially less NO_x production than if the system were operated with a single stage at the same overall equivalence ratio (McKinney et al., 2007). Specifically, the work conducted by Rocha et al. (2021) simulated the performance of RQL combustor at global equivalence ratio 0.4 and a pressure at 20 atm. They found at 20 atm, the combustor could still be stable by examining the flame speeds of ammonia at those conditions. High H_2 emission (over 3000 ppm) was observed in the exhaust even though the NO_x emission could be as low as approximately 50 ppm. However, it is still unknown how and at what operating conditions (equivalence ratio, pressure and residence time) to minimize NO_x , nor what NO_x levels are achievable. It is also concerning on the high H_2 level in the exhaust in RQL combustor and not clear how should H_2 be burnt out while maintaining NO_x level low.

A useful benchmark and guideline to have for ammonia combustor design is the minimum theoretical NO_x emissions that are achievable under ideal conditions (perfectly premixed limit, no heat loss). In our recent study (Gubbi et al., 2023), we analyzed the feasibility of ammonia combustion from NO_x emission perspective. In this work, we aim to establish the theoretical minimum NO_x and conducted detailed analysis for pure ammonia combustion. In other words, the objective of this work is not to simulate a given combustion system, but rather the fundamental minima that can be achieved via technology development. This approach mirrors that done by Goh et al. (2019) for lean-lean, axially fuel staged combustors. This work will provide theoretical guidance on practical combustor design

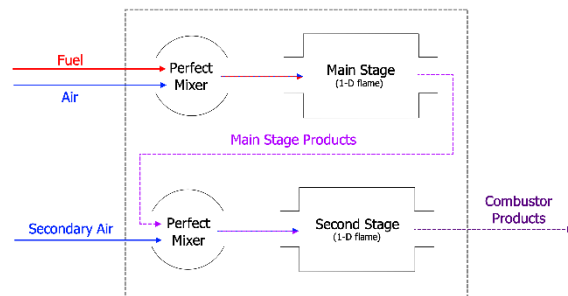


Figure 1: Schematic of the staged combustor reactor network model

for ammonia combustion to achieve minimized NO_x emissions employing staged combustor strategy. This is done over a range of prescribed combustor exit temperatures, combustor pressures, and combustor residence times with a constraint to ensure that the fuel is nearly all consumed.

The rest of the paper is organized as follows: Section 2 will discuss the reactor network model and kinetic model used in this study, as well as the reasoning behind the H_2 emissions constraint and the structure of the NO_x minimization problem. Section 3 will outline the results of general sensitivities, while Section 4 will discuss the results of optimization for a broad range of operating conditions.

MATERIALS AND METHODS

Reactor Network Model

Reduced order modeling by way of a chemical reaction network (CRN) was used for the minimum NO_x calculation. The reactor network used in this study is shown in Fig. 1. The reduced order modeling approach allows the main stage and second stage to be split and modeled as separate reaction networks. The reactor network modeling assumes perfect mixing and 1-D flame inside each stage. Adiabatic, constant pressure batch reactors were used to mix various streams of gas coming into each stage (nonreacting). For the main stage, streams of fuel and oxidizer were mixed, while for the second stage, the products from the main stage were mixed with secondary air. The mass flow rates for each stream were calculated based on the global and main stage equivalence ratios (Φ_{global} and Φ_{main}). The global equivalence ratio is specified, based upon the target combustor exit temperature.

The free flame model present in Cantera (Goodwin et al., 2022) outputs solutions in spatial coordinates, so numerical integration of axial velocities and distances between grid points was necessary to convert the solution to temporal coordinates. Although the flame zone generally starts at the same point in spatial coordinates, variations in flame speeds at different operating conditions cause variations in starting times for the flame. For this reason,

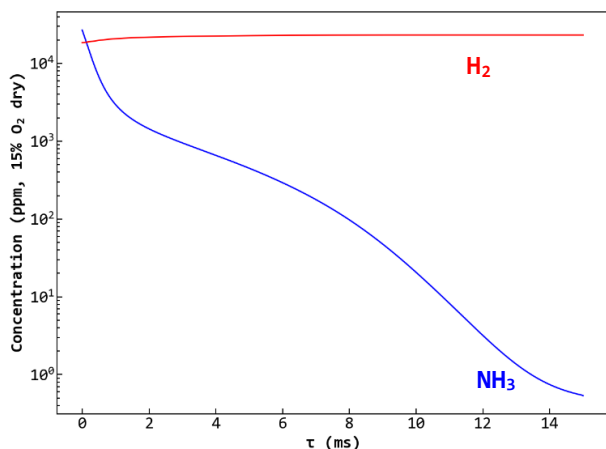


Figure 2: NH_3 & H_2 evolution in a rich ammonia-air ($\Phi = 1.2$, $P = 1$ bar)

peak NH_2 concentration was used to define the start of the flame zone. As one of the first species in the ammonia-air reaction pathway (Alnasif et al., 2022), it is a useful baseline for referencing the rest of the system in temporal coordinates.

All calculations were done using Cantera (Goodwin et al., 2022), which is an open-source library that can simulate chemical kinetics problems. The kinetic model used in this study is from Mei et al. (2019). In the simulation, fuel was pure ammonia, with a fixed temperature of 300 K. Oxidizer was synthetic air (79% N_2 and 21% O_2), with a preheat temperature of 650 K. Optimization was achieved at fixed values of Φ_{global} (overall equivalence ratio which controls combustor firing temperature), combustor pressure, and τ_{global} (total residence time) for a specific case. Since the simulation is under ideal conditions, NO_x is dominantly NO, and NO_2 and N_2O emission is negligible therefore NO is used instead of NO_x in the following discussion. Each of these parameters were also individually varied to study their effects on minimum NO, which will be discussed in later sections.

Optimization Problems and Constraints

In the prior study of Goh et al. (2019), which determined minimum NO_x levels and fuel splits for lean-lean, natural gas fired systems, a constraint on combustor exit CO was used. This was needed, because otherwise the optimizer would simply inject all secondary fuel immediately at the combustor exit, which leads to the minimum NO_x solution but incomplete combustion with high CO emission. An analogous procedure was used here. However, rather than constraining NH_3 , H_2 is needed to be constrained. The reason for this constraint is that in the main stage, a major product of rich NH_3 flame is H_2 owing to insufficient O_2 , as illustrated in Fig. 2. In Fig. 2, time zero starts to count from where NH_2 peaks to ensure consistent definition of residence time. NH_3 levels themselves are in the parts per billion level because of fast NH_3 decomposition at flame temperatures. Indeed, if NH_3

levels were used as the constraint, it is very possible to have significant levels of unburned fuel in the form of H_2 , while having essentially zero NH_3 emissions. Sufficient residence time is required in the second stage to oxidize H_2 , even while such condition promotes NO_x formation via the Zeldovich mechanism and other routes. Therefore, we utilized an exit H_2 constraint of 1.25 times its equilibrium value at the combustor exit temperature.

The optimization can be approached as minimization problem of the following form:

$$\begin{aligned} &\text{Minimize: } \text{NO}(\mathbf{x}) \\ &\text{with constraint: } \text{H}_2(\mathbf{x}) \leq 1.25 * \text{H}_{2\text{eq}}(\Phi_{\text{global}}) \end{aligned} \quad (1)$$

The variable \mathbf{x} is a vector consisting of design variables Φ_{main} and τ_{sec} (second stage residence time). $\text{H}_{2\text{eq}}$ is the equilibrium H_2 concentration at given equivalence ratio, τ_{global} . The coming sections of this paper will showcase the inverse relationship between NO and H_2 with regards to τ_{sec} (a lower value will mean lower NO level, but a higher value will allow for more relaxation of H_2). Generally speaking, it is favorable to keep τ_{sec} low to limit NO, however, sufficient time is needed to oxidize H_2 .

RESULTS AND DISCUSSION – GENERAL SENSITIVITIES

This section presents general results, illustrating the dependence of exit NO on various combustor design parameters. Results and discussion of optimized configurations are shown in Sec. 4. All results are displayed only for NO as it is the dominant species contributing to NO_x emissions in ammonia combustion. All emissions are specified in nanograms per Joule of energy input (ng/J) and parts per million (15% O_2 dry).

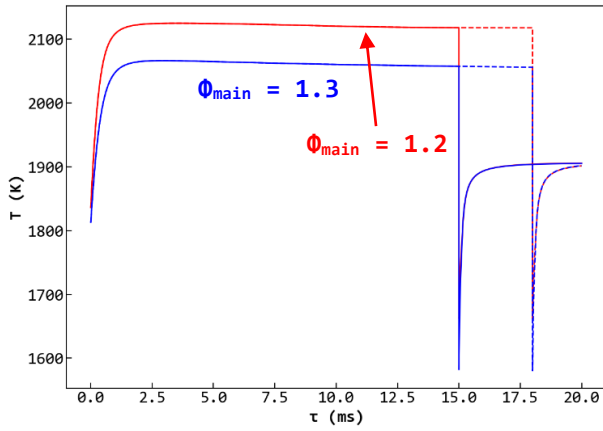
NO Dependence on Φ_{main} and τ_{sec}

At a given Φ_{global} (0.672) and total residence time of the entire combustor (20 ms), Fig. 3 shows the temperature profile and NO evolution through the combustor (main stage and second stage) at different Φ_{main} and τ_{sec} for several non-optimum configurations to illustrate basic sensitivities. Figure 3 showcases four different combustor configurations which come from all possible combinations of two Φ_{main} and two τ_{sec} values. While the global parameters for the combustor are fixed, with a firing temperature of 1900 K, the NO emissions at the exit are substantially different between configurations. These results demonstrate the minimization problem of NO by varying these two parameters.

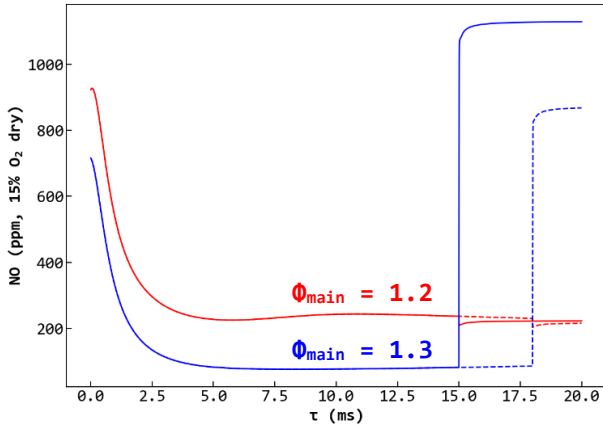
In Fig. 3, the sudden changes in the y-value coincide with the secondary stage where air is injected, which drops the temperature of the main stage products and also increases the total mass within the system.

Main Stage Equivalence Ratio Dependence

Figure 4 shows the variation of NO and H_2 levels at the combustor exit as Φ_{main} is varied while Φ_{global} and τ_{sec} are



(a) Temperature Profile



(b) NO Evolution

Figure 3: (a) Temperature profile and (b) NO evolution through combustor (solid line: $\tau_{\text{sec}} = 5$ ms, dashed line: $\tau_{\text{sec}} = 2$ ms; $P = 1$ bar)

fixed. NO emission is minimized at approximately $\Phi_{\text{main}} = 1.22$, with this optimum value varying slightly with conditions, such as firing temperature, which will be discussed later. The very small range of H_2 emissions indicates that H_2 oxidation can be completed in less than 5 ms for this condition.

Second Stage Residence Time Dependence

The variation of NO and H_2 levels at the combustor exit as a function of τ_{sec} is shown in Fig. 5 at fixed global residence time and $\Phi_{\text{main}} = 1.22$. In this simulation, the change of τ_{sec} is accompanied by a change in value of τ_{main} accordingly. A monotonic trend can be observed in emissions, where NO and H_2 have an inverse relationship when varying τ_{sec} . Figure 5(a) shows that longer second stage residence time will cause NO levels to rise owing to NO formation through Zeldovich mechanism. Moreover, higher exit temperature increases NO emission moderately. At the 1600 K condition ($\Phi_{\text{global}} = 0.459$), more air is injected in the second stage, thus creating significant abundant O radicals which are then consumed by various nitrogen-carrying species (such as NH and HNO) to

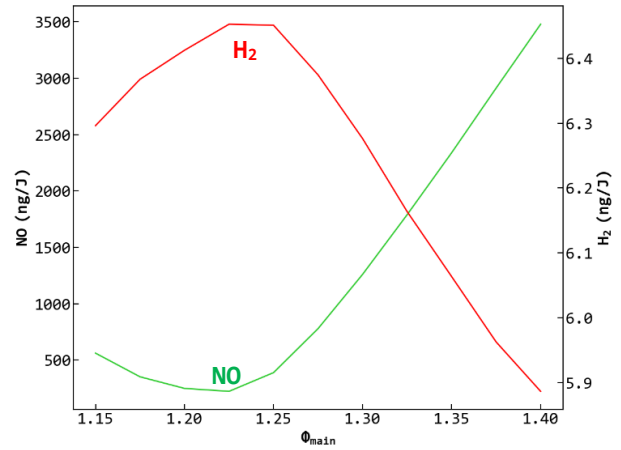
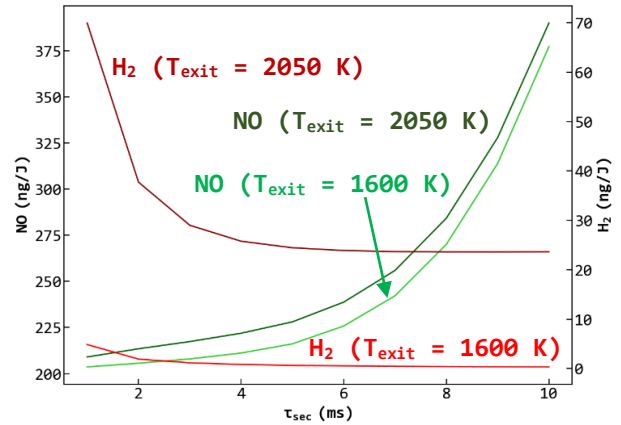
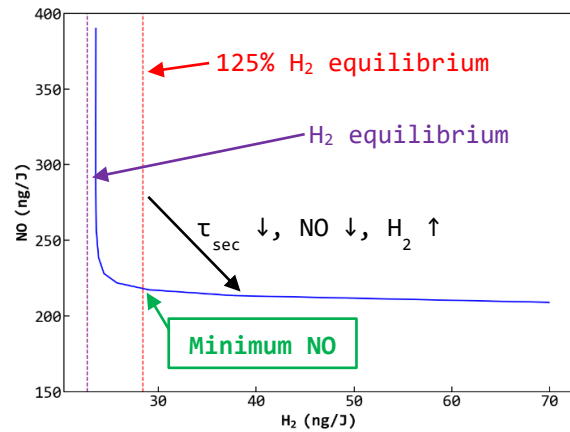


Figure 4: Variation of NO and H_2 emissions with Φ_{main} for a fixed $T_{\text{exit}} = 1900$ K and $\tau_{\text{sec}} = 5$ ms



(a) NO and H_2 vs. τ_{sec} (ms)



(b) NO vs. H_2

Figure 5: (a) Variation of NO and H_2 emissions with τ_{sec} and (b) relationship between NO and H_2 for a fixed $\Phi_{\text{main}} = 1.22$

produce NO. At higher temperatures, such as 2050 K ($\Phi_{\text{global}} = 0.8$), less O radicals are present even the Zeldovich mechanism becomes much more significant in NO contribution. Clearly, this τ_{sec} sensitivity in the secondary

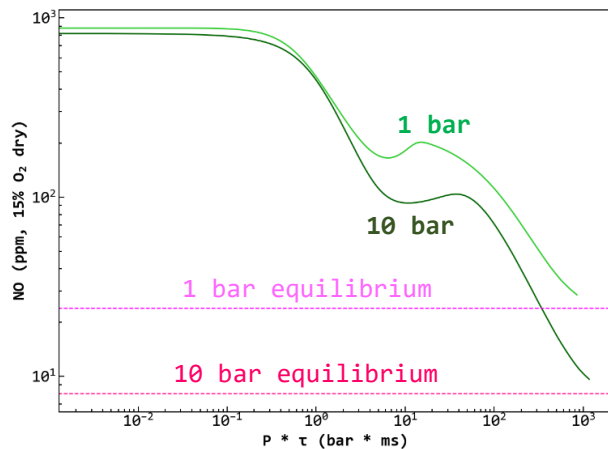


Figure 6: Main stage NO evolution to 125% equilibrium level ($\Phi = 1.22$)

stage is a strong function of the exit temperature and equivalence ratio. Conversely, higher τ_{sec} will allow for more complete combustion of H_2 , although there are diminishing returns as the H_2 approaches equilibrium.

Figure 5(b) shows the relationship between NO and H_2 emissions using results for the 2050 K condition from Fig. 5(a), with residence time varying along the curve explicitly illustrating the trade off in emissions between the two species. Also indicated in the figure is the criteria that H_2 emission is less than or equal to 125% of the equilibrium H_2 level based on the global equivalence ratio. The equilibrium and constraining lines are indicated in Fig. 5(b) by purple and red dashed lines, respectively, from which the defined minimum NO can be observed. The decreasing residence time while moving down the curve is also noted, meaning that the optimum τ_{sec} , will be the smallest possible residence time that allows for sufficient oxidation of H_2 .

Analysis of NO Production Sources

It is helpful to look at the individual NO contributions between the main and secondary stages. Main stage NO can be broken down even further into an equilibrium contribution (the minimum possible NO from the main stage) and some extra amount of “unrelaxed” NO. The resulting decomposition is as follows:

$$NO_{total} = NO_{main,eq} + NO_{main,unrelaxed} + NO_{second} \quad (2)$$

The NO evolution for a rich ammonia flame is shown in Fig. 6. Note that long residence times are required for rich NH_3 /air mixture to reach equilibrium, with this relaxation time dropping with pressure. Also note that we have scaled the x-axis with pressure to readily illustrate both pressure results together. Although the residence times required to reach equilibrium may not be feasible for practical

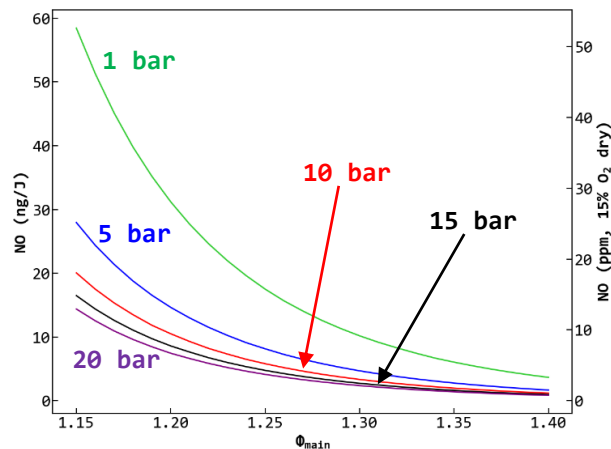


Figure 7: Main stage equilibrium NO contribution over a range of equivalence ratios

combustors, it still could be a significant factor to be considered as longer main stages can drastically reduce NO emissions.

Figure 7 plots an equilibrium calculation, which quantifies $NO_{eq,main}$ as a function of equivalence ratio and pressure. High pressure and richer main stage conditions are favored for lower NO_x emission, as expected. These calculations suggest that if an ammonia combustor is designed that allows the main stage to reach equilibrium, and minimizes NO production in the secondary stage, NO emissions as low 11 ng/J (10 ppm) can be achieved.

RESULTS AND DISCUSSION – OPTIMUM CONFIGURATIONS AND MINIMUM NO

Having illustrated basic sensitivities to key parameters, this section presents results on optimum configurations and values.

Combustor Firing Temperature Sensitivities

Figure 8 plots minimum NO_x levels over a range of firing temperatures from 1600 to 2050 K for a 20 ms combustor residence time at 1 bar. We limited the residence time to 20 ms in this simulation for the sake of existing combustor technologies. Residence time certainly be extended in simulation to achieve even lower emission levels. The different NO contribution bars are also shown. The bottom curve represents the NO level if the main stage operated until it reached equilibrium. Results indicate that higher emissions are favored at lower firing temperature, and NO emissions under 250 ng/J (225 ppm) are achievable at higher firing temperature. The majority of contribution is from unrelaxed NO in the main stage, indicating that even lower emissions can be achieved if the combustor residence time is increased.

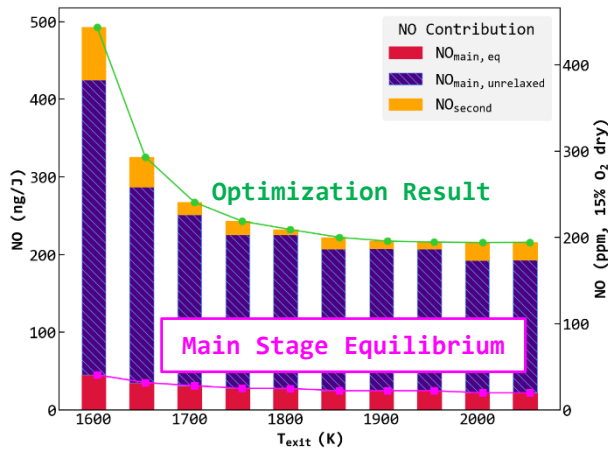
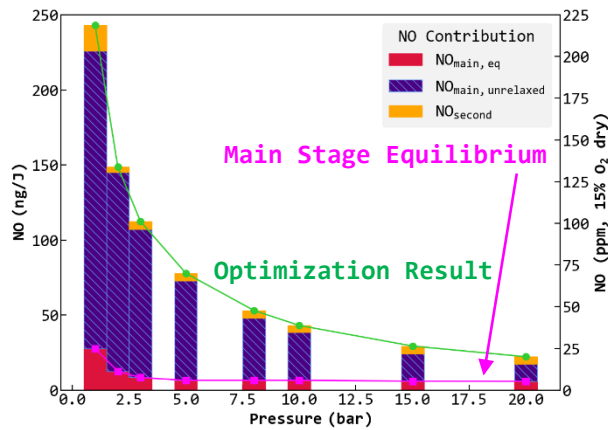
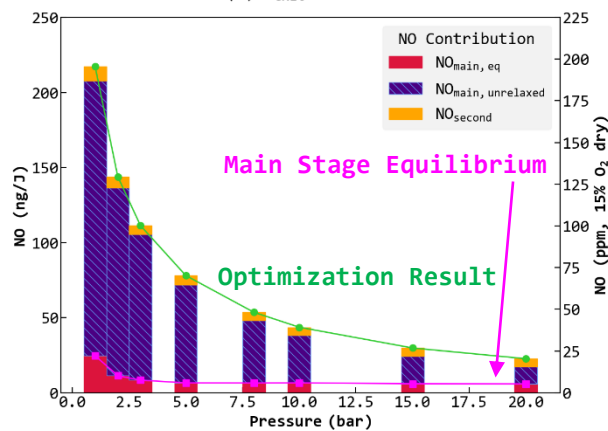


Figure 8: Minimum NO at various firing temperatures ($P = 1$ bar)



(a) $T_{\text{exit}} = 1750$ K

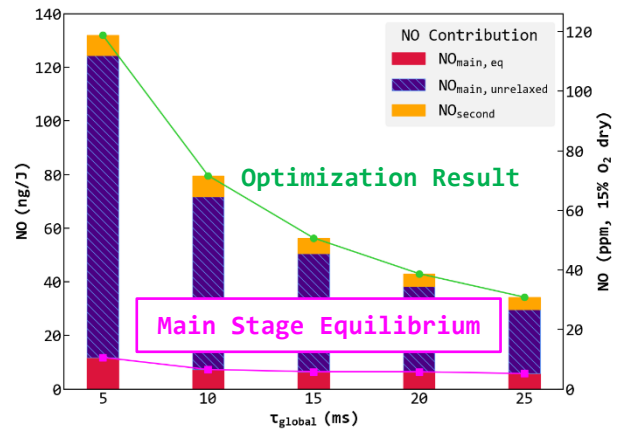


(b) $T_{\text{exit}} = 1900$ K

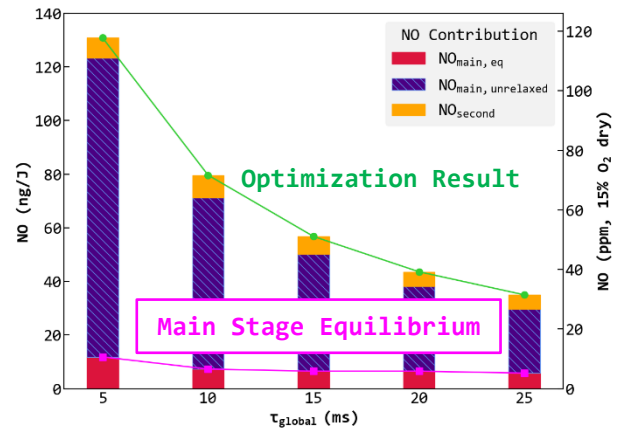
Figure 9: Minimum NO at various combustor pressures (a) $T_{\text{exit}} = 1750$ K (b) $T_{\text{exit}} = 1900$ K

Combustor Pressure Sensitivities

The combustor pressure was also varied from 1 bar up to 20 bar for a combustor with total 20 ms residence time and optimized to examine the pressure effects on minimum



(a) $T_{\text{exit}} = 1750$ K



(b) $T_{\text{exit}} = 1900$ K

Figure 10: Minimum NO at various global residence times (a) $T_{\text{exit}} = 1750$ K (b) $T_{\text{exit}} = 1900$ K ($P = 10$ bar)

NO. The results are shown in Fig. 9(a) and Fig. 9(b) at two different combustor exit temperatures (with corresponding global equivalence ratios of 0.559 and 0.672). Although the difference in NO was large at low pressure conditions, this difference diminished as the pressure was increased, and nearly identical emissions are achieved for high pressure regardless of the firing temperature. High pressure also contributes to faster approach to equilibrium at the main stage, as noted previously, and this decrease in the unrelaxed NO contribution is significant. For high pressure conditions, the model predicts emissions could be as low as 25 ng/J (23 ppm).

Global Residence Time Sensitivities

The effect of global residence time was also investigated, which is especially critical when considering the main stage unrelaxed NO contribution. A range of 5 ms to 25 ms was studied operating at 10 bar, with two temperature conditions analyzed similar to the pressure sweep case (Fig. 10). As expected, the minimum achievable NO decreases for longer combustors, and the main stage unrelaxed NO contribution is lower for longer residence times.

CONCLUSIONS

In this work, reduced order modeling was conducted to investigate the theoretical minimum NO_x for ammonia combustion in a staged combustor with rich burn main stage. The rich burn main stage could reduce NO_x emission comparing to stoichiometric and lean flames, therefore reduce the overall NO_x emission. However, rich burn of ammonia produces significant amount of H₂ owing to NH₃ decomposition in the rich flame zone. The second stage needs to be sufficiently long to oxidize H₂ while NO_x is produced too simultaneously. Optimization of such combustion strategy by constraining H₂ emission while minimize NO_x emission shows that lower NO_x emission could be achieved with increased pressure and longer residence time of the main stage to accelerate the rich main stage to approach equilibrium which yields the lowest amount of NO_x. The dominate NO_x in the exhaust is majorly from the unburnt NO produced in the rich main stage. Given conditions which are relevant to existing gas turbines (20 atm, 1900 K firing temperature and 20 ms combustor residence time), the theoretical minimum NO_x in a stage combustor could be lower than 25 ng/J (23 ppm).

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