A Kinetics Study on the NOx Emissions of Axially Staged Combustion System for Gas Turbine Applications

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ABSTRACT
Axially-staged combustor has great NOx-abatement potential for advanced gas turbines of high turbine inlet temperature in the future. A chemical reactor network (CRN) model is established for the axially-staged combustor. The effects of important combustor parameters on NOx emissions under operating conditions of advanced gas turbine were simulated using the CRN model with CHEMKIN. Simulation results are first presented for perfectly mixed combustion systems to study the effect of the first stage temperature, residence time split, and flow rate split on NOx emissions. It is found that staged combustion can significantly reduce NOx emissions compared to single stage combustion under lean premixed conditions. It is also shown that (a) the first stage temperature and residence time split have a greater impact on NOx emissions than flow rate split and (b) NOx emissions can be reduced significantly by shortening residence time of high temperature stage or decreasing the first stage temperature. Then, the effect of unmixedness at the inlet of the second stage is introduced to model the axially-stage combustor. It is worth noting that the mixing between fuel and air which are injected into the second stage chamber has a greater impact on NOx emissions than the mixing between the exhaust gas from upstream and the fresh-mixture jet of the second stage.

INTRODUCTION
Demands for higher-efficiency gas turbines are pushing the turbine inlet temperature (TIT) to material limit, which can result in significant increase in NOx emissions that conflict with the increasingly-strict environmental regulations. For example, the MHI J-class gas turbine has a NOx emission of 25 ppm at a TIT of 1600 °C, exceeding the emissions limits of most of the developed countries. The continued interest in advancing the efficiency of gas turbines will likely keep driving the increase in the TIT, and hence the increase in NOx emissions. Lieuwen et al. [1] predicted that at future gas turbine operating conditions, significant NOx emissions may be produced if conventional lean premixed combustion system is to be used. Therefore, it is necessary to develop advanced combustion technologies to meet the regulatory requirements. Staged combustion is a promising technology that can significantly reduce the NOx emissions while offering flexibility in load-variation of the combustor.

Figure 1 illustrates the basic concept of an axially-staged combustor. The combustor consists of two combustion stages. The compressor discharge air is split into primarily two parts: the first part is fed into the first stage (stage 1), while the other part is fed into the second stage (stage 2). Fuel is also split into two parts, and fed into the two stages separately after mixing with the air of each stage. Staged combustion offers the flexibility of varying the combustor load by adjusting the fuel split into the two stages, while operating both of the stages at lean premixed combustion mode to meet emission standards. At low load, most (or all) of the fuel is fed into stage 1, while above a certain load more fuel is supplied to stage 2.

\[
\begin{align*}
Q_1 &= Q_{1_{\text{air}}} + Q_{1_{\text{fuel}}} \\
Q_2 &= Q_{2_{\text{air}}} + Q_{2_{\text{fuel}}} \\
Q_{1_{\text{air}}} &= T_{\text{inlet}} \cdot \tau_1 \\
Q_{2_{\text{air}}} &= T_{\text{inlet}} \cdot \tau_2
\end{align*}
\]

\(Q_{1_{\text{air}}}, Q_{2_{\text{air}}}:\) the mass flow of air in both the stages  
\(Q_{1_{\text{fuel}}}, Q_{2_{\text{fuel}}}:\) the fuel mass flow in both the stages  
\(T_{\text{inlet}}, T_{\text{outlet}}:\) the inlet and outlet temperatures, \(\tau_1, \tau_2: \) the residence time of each stage.

Figure 1: Axially-staged combustor concept. \(T_{\text{inlet}}: \) the inlet temperature, \(T_{\text{outlet}}: \) the outlet temperature, \(\tau: \) the residence time, \(Q: \) the mass flow.

Therefore, staged combustion has gained interest for the research and development of future advanced gas turbines. Sattelmayer et al. [2] stated that the NOx reduction potential of staged combustion is low unless mixing between the stages takes place before the secondary combustion. Hayashi and Himada [3] proposed a two-stage combustor concept for achieving stable combustion with ultra-low NOx emissions for a wide range of operating conditions at atmospheric pressure. In their study, the air-flow of air in both the stages and the fuel-flow rate in stage 1 were fixed. Then, only the secondary fuel flow rate were changed to cover the range of fuel-air ratios utilized in a gas turbine. Hayashi et al. [4] extended their previous work and investigated the NOx...
emissions of kerosene-fuel combustion in the two-stage combustor at 600 K inlet air temperature and 0.8 MPa pressure for two sizes of mixture injection tubes. Ahrens et al [5-7] have demonstrated that staged combustion has the potential for low NOx at high pressure and the mixing between the exhaust gas from upstream and the fuel-air mixture jet of stage 2 has an impact on NOx emissions. Winkler et al [8] compared three kinds of mixing devices between the stage 1 exhaust gas and the stage 2 fresh mixture. Mixing devices not only need to mix gases efficiently and compactly, but also ensure the stability of stage 2 flame. The effects of these devices on emission reduction are as good as each other.

A key feature of the axially-staged combustion is that the air/fuel flow split and the residence time split between the two stages can be adjusted to achieve optimized operation in terms of emissions and lean blow off. In the previous studies, however, these important parameters were fixed. Effects of these splits on NOx emissions were not well studied under operating conditions of gas turbines. Moreover, the unmixedness between fuel and air at the exit of the stage 2 burner has not been taken into account. Understanding these issues is essential for the design and operation of axially-staged combustors to meet emissions requirements.

In light of the knowledge gap, we present a kinetics study on the NOx emissions of an axially-staged combustor upon variation of key design parameters, including the stage 1 temperature, flow rate split and residence time split between stage 1 and stage 2. The axially-staged combustor was simulated using CRN model in the CHEMKIN software. This paper will first focus on the perfectly premixed combustion system, and then introduce unmixedness at the stage 2 to model the practical combustion system.

**METHODOLOGY**

**Modelling staged combustion for perfectly premixed combustion system**

Figure 2 shows the CRN model of the staged combustor under perfectly mixed conditions. In this model, perfectly stirred reactor (PSR) is used to simulate the recirculating flame zone and plug flow reactor (PFR) is used to simulate the post flame zone, following the approach of Li et al. [9]. The upstream PSR-PFR is the model of the first stage chamber and the downstream PSR-PFR is the model of the second stage. The overall residence time of this CRN model is 15ms, and the residence times of the two PSRs are both set to 1ms, which are typical for industrial gas turbines [9]. The residence time split between the two stages is varied by changing the volumes of the two PFRs when mass flow rate at the inlet of stage 1 and stage 2 is constant. The flow rate split between the two stages is set to three conditions \(k = 0.5, 1, 2\).

![Figure 2: Schematic of perfectly premixed model](image)

The effects of the stage 1 temperature, residence time split, and flow rate split on NOx emissions was studied systematically using this model. The simulations are performed using the CHEMKIN-PRO software package with GRI-Mech 3.0. Typical natural gas composition (94.22% CH4, 15% C2H6, 2.62% N2) is used for the simulations [9]. The inlet temperature of compressed air is 700K, and the combustor pressure is 25atm.

**Definition of unmixedness**

Under realistic conditions, reactants cannot be mixed perfectly in combustors. As shown in Figure 3, the fuel and air are mixed in the stage 2 burner before being injected in the form of jets. The injected fuel-air mixture jet then mix and react with the crossflow of the exhaust gas from the first stage. Two types of unmixedness are defined, the fuel-air unmixedness and the jet-in-crossflow unmixedness, to quantify the mixing at the inlet of stage 2. The Jet-in-crossflow unmixedness quantifies the mixing between the exhaust gas from the first stage and the fresh fuel-air mixture jet of the second stage, while the fuel-air unmixedness quantifies the mixing of fuel-air at the exit of the stage 2 burner. This paper assumes that two types of unmixedness do not interfere with each other.

![Figure 3: Conceptual illustration of the two types of Mixing at the inlet of stage 2](image)

A typical approach for quantifying the fuel-air unmixedness in a combustor is to assume that the mixture can be approximated by a normal distribution about some mean fuel mass fraction[10]. The distribution is then defined by an unmixedness parameter \(u_1\), where

\[
u_t = \sqrt{\frac{\sigma^2}{\bar{Y}_f(1-\bar{Y}_f)}}, \tag{1}\]

and \(\sigma\) is the standard deviation of the distribution and \(\bar{Y}_f\) is the mean fuel mass fraction.
Similarly, the jet-in-crossflow unmixedness can be defined as the following formulation:

\[ u_z = \sqrt{\frac{\sigma^2}{\bar{Y}_f(1-\bar{Y}_f)}} , \]  

(2)

where \( \sigma \) is the standard deviation and \( \bar{Y}_f \) is the mean jet mass fraction of fresh mixture jet which can be calculated by \( \bar{Y}_f = \frac{Q_1}{(Q_1 + Q_2)} \). However, this jet mass fraction cannot be assumed as a normal distribution. By fitting the data of jet mass fraction which is measured in the experiments of Hoferichter et al. [6],the distribution of jet mass fraction are approximated by the beta distribution as shown in Figure 4.

**Figure 4: real, beta and normal distribution**

**Modelling Staged combustion considering unmixedness**

For simplicity, it is assumed that the unmixedness only exists in flame zone of stage 2 and gases are perfectly mixed in post flame zone. The flame zone is modelled as a network of parallel PSRs as shown in Figure 5.

**Figure 5: Schematic of partially mixed model**

Each PSR in the flame zone model represents a fluid micelle with different ratio of fuel to air or jet to cross flow. The principle to determine the number of parallel PSRs is that NOx emissions do not change with the number of PSRs increasing. The change of NOx emissions is calculated as

\[ \Delta_{NOx}(i+1) = \frac{NOx(i+1) - NOx(i)}{NOx(i)} , \]  

(3)

where \( i \) refers to the number of parallel PSRs. With the curve of probability density function being gentler, more and more PSRs are required. The results show that models of normal distribution and beta distribution both need at least nine PSRs.

**RESULTS AND DISCUSSION**

**Effect of the stage 1 temperature for perfectly mixed combustion system**

The effect of fuel distribution on NOx emissions is equivalent to the effect of the stage 1 temperature on NOx emissions, when the air flow of stage 1 and stage 2 is constant.

Figure 6 shows the effect of the stage 1 temperature on NOx emissions. The dash line in Figure 6 is for the baseline case with conventional single stage combustion. When combustor outlet temperature (COT) is over 1850 K, staged combustion has greater potential for NOx reduction than single stage combustion. NOx emissions decrease with the stage 1 temperature dropping as the COT is constant, but it is not obvious at low temperature of stage 1 (\( T_1<1700 \) K).

In Figure 6, the fact that there is an intersection between the dash curve and the solid curve (\( T_1=1800 \) K) might be unusual at first look. The residence time of the jet in the stage 2 chamber is shorter than the baseline. Thus, NOx emissions of staged combustion are lower than the baseline at the right side of the intersection. However, the stage 1 temperature is higher than the corresponding temperature of the single stage combustor when combustor outlet temperature is higher than the stage 1 temperature therefore it results in more NOx produced in staged combustion at left side of the intersection. In summary, the stage 1 temperature cannot exceed combustor outlet temperature while designing low-emission staged combustion combustor.

**Figure 6: Effect of temperature on NOx emissions.**

Conditions: \( P=25 \) atm, \( T_{pre}=700 \) K, \( \zeta=10 \) ms, \( k=Q_j/Q_s=2 \). Baseline is single stage combustion under the same conditions.
Figure 7 shows the ratio of stage 1 NOx to total NOx as a function of COT. The ratio is defined as

$$C_1 = \frac{N_1 Q_{1,1}}{N_1 (Q_{1,1} + Q_{2,1})},$$

where $N$ [ppm] is the NOx emission. $Q$ is the bulk flow and subscript 1&2 represents stage 1 & stage 2 respectively. As shown in Figure 7, the ratio is decreasing with the stage 1 temperature dropping. Therefore, NOx emissions mainly stem from second stage and the effect of the stage 1 temperature on NOx emission is continually weakened with the stage 1 temperature dropping.

![Figure 7: The ratio of the stage 1 NOx to total NOx](image)

**Effect of flow rate split for perfectly mixed combustion system**

Figure 9 shows the effect of flow rate split on NOx emissions for $T_1 = 1700$ K, 1800 K and 1900 K. The NOx emissions decrease with reducing flow-rate-split scalar ($k$) at the stage 1 temperature of 1900K, while the sensitivity of flow rate split on NOx emissions is low at the stage 1 flame temperature of 1700K and 1800K. The latter seems to be contrary to the fact that NOx emissions increase with residence time. It might be explained by lots of stage 2 prompt NOx decreasing with the reduction of the stage 2 flow rate. Hoferichter et al [6] found that the region where the stage 2 jet is mixed with the hot cross flow produces a large amount of prompt NOx. Thus, the increase of NOx caused by adding more fuel-air mixture into stage 1 is almost offset by the decrease of prompt NOx at the inlet of the stage 2. However, NOx increase cannot be completely offset at the stage 1 temperature of 1900K. The high temperature of stage 1 results in lots of post flame NOx, which is too much to be counteracted completely.

![Figure 8: Effect of the stage 1 temperature on NOx emissions. $T_{outlet} = 1985$ K](image)

According to the above analysis, as much fuel as possible should be injected to stage 2 to reduce the NOx emissions temperature when the COT is constant. However, the effect of the stage 1 temperature on NOx emissions is less and less obvious with the stage 1 temperature decreasing. Figure 8 is the relationship between the NOx emission and the stage 1 temperature at a COT of 1985 K. As shown in Figure 8, the minimum NOx emission is 19 ppm apparently that does not meeting the emission standard. When the stage 1 temperature is 1725 K the NOx emission is 1% larger than the minimum value (19ppm). Continuing to reduce the stage 1 temperatures has a little effect on NOx reduction at a high COT, and hence it is necessary to reduce the NOx emission by adjusting flow rate split and residence time split.
Effect of residence time split for perfectly mixed combustion system

Figure 11 illustrates the effect of residence time split on NOx emissions at different stage 1 temperature. As shown in the three subgraphs, NOx emissions decrease significantly with the stage 1 residence time shortened at high COT \( T_{\text{outlet}} > 1850 \text{ K} \), while the residence time split has a little effect on NOx emissions when the COT is low. That is because NOx generation in stage 2 is mainly post flame NOx at high COT, while prompt NOx accounts for a larger proportion in stage 1 and 2 at low COT. Since post flame NOx increases with the residence time but residence time has a little effect on prompt NOx, the residence time split has greater impact on NOx emissions at high COT. Therefore, with the continuous increase of the outlet temperature in future, the residence time split is becoming more and more important for reducing NOx emissions. In other words, shortening the residence time of high temperature combustion section (the stage 2 chamber), has a significant effect on NOx reduction.

Figure 12 illustrates the influence of residence time split on NOx emissions of three kinds of flow rate split. The trends that NOx emissions are decreasing with the stage 2 residence time in the three graphs are almost as same as each other. It is also worth noting that the sensitivity of flow rate split is always low no matter how residence time split changes. Compared with the Figure 9a, the effect of residence time split on NOx emissions in staged combustion is greater than the flow rate split. With the COT rising in future, the residence time split in staged combustor plays a more and more important role.
Figure 11: Effect of residence time split for $T_1$=1600 K, 1700 K and 1800 K. Flow-rate-split scalar $k$ = 2.

Figure 12: Effect of residence time split for $k$ = 0.5, 1 and 2. Stage 1 flame temperature $T_1$ = 1700 K.

Effect of the fuel-air mixing at the exit of the stage 2 burner

In order to study the effect of the fuel-air unmixedness, it is assumed that the stage 2 fuel-air mixture is perfectly mixed with the stage 1 exhaust gas and gases in stage 1 are completely mixed. The ratio of the stage 1 flow rate to the stage 2 flow rate in parallel PSRs is equal to each other

$$\frac{Q_{1,i}}{Q_{2,i}} = k,$$

(5)

where $Q$ is the mass flow, subscript 1&2 represents stage 1 & stage 2 respectively, subscript $i$ is the parallel PSRs label, $k$ is the flow rate split scalar.

Figure 13 illustrates the relationship between the stage 2 fuel-air unmixedness and NOx emissions under the two types of flow rate split. The NOx emissions of two graphs both increase with unmixedness, but the increase of NOx emissions in graph (a) is greater than in graph (b). That is because the proportion of NOx emissions resulted by unmixedness increases as the stage 2 fuel-air mixture flow increases. The effect of the unmixedness significantly increases with the COT rising. The reason is that the ratio of the stage 2 NOx to total NOx increases with the COT rising as shown in Figure 7. In summary, NOx emissions can be reduced by decreasing the flow rate of the stage 2 jet.
Effect of the jet-in-crossflow unmixedness at the inlet of stage 2 combustor

Based on the above analysis, the beta distribution can approximately fit the mass fraction distribution of the stage 2 jet. The mass fraction distribution of other kinds of mixing devices remains to be verified due to the limit of public experimental data.

Figure 14 and Figure 15 show the effect of jet-in-crossflow unmixedness on NOx emissions. Both figures were obtained using CRN model under the conditions that shown in Table 1. The jet-in-crossflow unmixedness has a little effect on NOx emissions when jet-in-crossflow unmixedness is below 0.075, while the fuel-air unmixedness has greater impact on NOx. That is mainly because the fuel-air unmixedness is defined by the distribution of fuel mass fraction, while the jet-in-crossflow unmixedness is defined by the distribution of jet mass fraction. Since fuel flow accounts for a small fraction of the jet flow, the temperature gradient of the parallel PSRs is not great therefore the NOx emissions is not sensitive to jet-in-crossflow unmixedness.

As shown in Figure 14 and Figure 15, NOx emissions keep increasing with the jet-in-crossflow unmixedness being larger. When the COT is lower than 1750 K, the jet-in-crossflow unmixedness has a little effect on NOx emission. However the effect on NOx emissions increases sharply with the COT rising. If the unmixedness is too large it may result in more NOx emissions than the single stage combustion, thus the staged combustion loses its value of emission reduction. The jet-in-crossflow unmixedness of Hoferichter’s staged combustion combustor [6] is 0.41. The emission of the combustor still meet the standard at a COT of 1908 K, while it is far more than the standard at a COT of 2000 K. Therefore, it is very important to design a mixing device which maintains a low unmixedness in future.

CONCLUSIONS

This paper has demonstrated that the staged combustion technology has the potential to significantly reduce NOx emissions. Effects of the stage 1 temperature, residence time split, flow rate split and unmixedness on NOx emissions were systematically studied using CRN model. Major conclusions are as follows:

1. The overall NOx emissions decrease with the stage 1 temperature. And the stage 1 temperature cannot exceed the combustor outlet temperature for the staged combustor to be effective in reducing NOx emissions.

2. Under perfectly mixed conditions, the stage 1 temperature and residence time split have greater impact on NOx emissions than the flow rate split. The key to reducing NOx emissions is to shorten the residence time of the high temperature section (stage 2) when the overall residence time is constant.
3. The impact of unmixedness on NOx emissions is greater with the flow rate increasing. It is effective to reduce NOx emissions by reducing the flow rate in the stage chamber where unmixedness is high.

4. The effect of the jet-in-crossflow unmixedness on the NOx emissions is much less than the effect of the fuel-air unmixedness on the NOx emissions. And the key for combustor design is to make fuel and air premixed perfectly in the stage 2 combustor.

**NOMENCLATURE**

- \( Q \): mass flow [g/s]
- \( Q_v \): bulk flow [cm³/s]
- \( T \): temperature [K]
- \( Y_f \): fuel mass fraction [-]
- \( Y_2 \): jet mass fraction [-]
- \( u_1 \): fuel-air unmixedness [-]
- \( u_2 \): jet-in-crossflow unmixedness [-]
- \( N \): NOX emissions [ppm]
- \( C_1 \): the ratio of the stage 1 NOx to total NOx [-]
- \( k \): flow-rate-split scalar [-]

**Greeks**

- \( \tau \): flow-rate-split scalar [-]
- \( \sigma \): standard deviation [-]
- \( \Delta \): change rate [%]
- \( \Phi \): equivalence ratio [-]

**Subscripts**

- 1: the first stage
- 2: the second stage
- i: the number of PSR

**Acronyms**

- TIT: turbine inlet temperature
- COT: combustor outlet temperature

**REFERENCES**


