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Kinetics Modeling on NO_x Emissions of Gas Turbine Combustors for Syngas Applications

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ABSTRACT

This paper presents a kinetics study on NO_x emissions of syngas gas turbine with RQL (rich-burn, quick-mix, lean-burn) combustion. The RQL combustor was simulated by a chemical reactor network (CRN) model using CHEMKIN-PRO program. The kinetic mechanism used in the simulation was developed by Zhang et al. (2017), dedicated to syngas fuel. NO_x emissions of RQL combustion were systematically studied under representative gas turbine operation conditions, and results show that RQL combustion significantly reduces NO_x emissions. Key parameters of RQL combustor, including air flow split and residence time split between rich and lean burn zones, were varied to investigate their effects on NO_x emissions. Analyses show that air flow split is the key factor determining NO_x formation.

Influences of mechanisms on NO_x prediction in the RQL combustor were also investigated. The GRI-Mech 3.0 mechanism was chosen for comparison. The syngas mechanism developed by Zhang et al. predicts lower overall NO_x emissions when the combustor outlet temperature is 1750K, and predicts higher overall NO_x emissions when the outlet temperature is 1908K. In the rich-burn zone of the RQL combustor, the syngas mechanism predicts lower NO_x production at 1750K, and almost the same NO_x production at 1908K compared with GRI-Mech 3.0. While in the lean-burn zone of the combustor, the syngas mechanism predicts higher NO_x formation at both 1750K and 1908K. Sensitivity analyses were conducted to find major reactions that influenced the NO_x prediction in each mechanisms. Results show that the dominating pathways of NO formation are not same in each mechanism. ROPs (rates of production) of these pathways were calculated to further explain the differences in predictions of each mechanism.

INTRODUCTION

Integrated gasification combined cycle (IGCC) is a promising solution for clean coal utilization by burning the

coal-derived syngas in a gas turbine. Due to flashback issues of hydrogen-rich syngas flames (Hui et al. 2007), current IGCC gas turbines usually use non-premixed combustors, which lead to higher NO_x emissions than lean premixed combustors. To meet the environmental regulations, alternative combustion methods need to be considered. RQL (rich-burn, quick-mix, lean-burn) combustion offers low NO_x emissions while having low risk of flashback. In RQL method, air was injected into the combustor separately, and the combustor was divided into rich-burn zone (with primary air and fuel injection), quick-mix zone, and lean-burn zone (with secondary air injection). RQL combustion can avoid the high temperature of stoichiometric flame and leads to less NO_x formation (Samuelsen, 2006).

RQL is widely used in both aero gas turbine and stationary gas turbine. (Pratt & Whitney used RQL in aero gas turbine (McKinney et al. 2007); Ingenito et al. (2014) conducted a numerical study on RQL combustor of a gas turbine used in aircrafts; General Electric developed RQL combustor for F-class stationary gas turbine (Feitelberg et al. 1998); Straub et al. (2005) investigated RQL stationary gas turbines combined with trapped vortex combustor.) In studies of RQL, the fuels are usually natural gas or hydrogen. Few studies focused on syngas. RQL combustor burning syngas still needs to be investigated.

Reaction mechanisms are also important in kinetics modelling of syngas RQL combustor. Sahu et al. (2014) conducted a detailed numerical study of NO_x kinetics in syngas flame. Five mechanisms were compared, showing that NO_x concentration predicted by each mechanisms are not same and major pathways that influence NO_x prediction in these mechanisms are different. However, Sahu's study was focused on opposed jet diffusion flame, which is far from RQL. Influences of different mechanisms on NO_x prediction in RQL combustor need to be studied. And this paper analysed the influences of two reaction mechanisms, including GRI-Mech 3.0 (Gregory et al. accessed 2017) and

a mechanism developed by Zhang et al. (2017) dedicated to syngas (named as ‘SYN-Mech’ for short in this paper).

This paper used chemical reactor network (CRN) in simulation. CRN is a widely used tool for combustor simulation. Hao (2014), Park et al. (2013) used CRN model to predict NO_x emissions in gas turbine, combined with computational fluid dynamics; Kroniger et al. (2017) used a simple CRN model to study the influences of hydrogen injection to gas turbine fuel.

REACTOR MODEL CONFIGURATION

Figure 1 shows the schematic of the CRN model for the IGCC gas turbine combustor. The model consists of a group of ideal reactors, including perfect stirred reactor (PSR), mixer and plug flow reactor (PFR). The PSR is a zero-dimensional reactor, which assumes that all reactants are perfectly mixed after entering the reactor. The mixer is a reactor that assuming no chemical reaction happens inside. The PFR is one-dimensional and assumes no axial difference but radial resolution of species, velocity, density, and temperature, etc. The rich-burn zone is simulated by a group of PSR. The quick-mix zone is simulated by a mixer. And the lean-burn zone is simulated by a PFR. All the fuel is injected into the rich-burn zone. And for the RQL combustor, the average equivalence ratio of the rich-burn zone should be always over one.

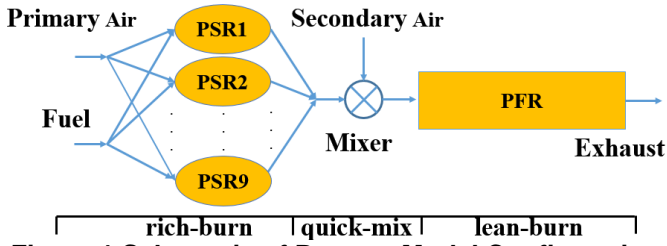


Figure 1 Schematic of Reactor Model Configuration

To simulate the mixing effect of fuel and air in the rich-burn zone, the equivalence ratio ϕ in the rich-burn zone is assumed to follow Gaussian distribution (Li et al. 2009) shown in equation (1). $\bar{\phi}$ stands for the average equivalence ratio in the rich-burn zone and σ stands for the standard deviation. And the mixing efficiency η is defined as equation (2).

$$f(\phi) = \frac{1}{\sqrt{2\pi}\sigma} \exp\left[-\frac{(\phi - \bar{\phi})^2}{\sigma^2}\right] \quad (1)$$

$$\eta = \left(1 - \sigma / \bar{\phi}\right) \quad (2)$$

The rich burn zone is separated to 9 PSRs and the equivalence ratio of the zone ranges from $(\bar{\phi} - 2\sigma)$ to $(\bar{\phi} + 2\sigma)$. The primary air injected into each PSR is determined by the probability distribution of the equivalence ratio within the PSR (calculated with MATLAB software). Fuel flowrates of each PSR are calculated from the air flowrates and average equivalence ratio of the PSR. The mixture of the secondary

air and the products of the rich-burn zone (PSRs) continue to react in the lean-burn zone (PFR).

The syngas used in the simulation has a composition of 25.7742% H₂, 56.993% CO, 15.22% N₂, 2.0128% CO₂. The preheat temperatures of both air and fuel are 600K. The pressure of the combustor is 12atm. Mixing efficiency η is set to 90%. The total residence time of the combustor is 20ms. Combustor outlet temperature, air flow split and residence time split are varied. The air flow split ratio is defined as secondary air flow rate divided by primary air flow rate.

Simulations were produced with the CHEMKIN-PRO software package. The mechanism used in the simulations was ‘SYN-Mech’. In addition, GRI-Mech 3.0 was also used for comparison to investigate the influences of different mechanisms on NO_x prediction.

RESUALT AND DISCUSSION

NO_x emissions of RQL combustor

Figure 2 shows the NO_x emissions at different combustor outlet temperature. Under RQL condition, air flow split ratio is set to 4 (20% primary air and 80% secondary air). Residence time of PSR is set to 5ms and residence time of PFR is set to 15ms. In this paper, NO_x emissions are corrected to 15% O₂ and dry condition if not specially mentioned. The base line is made for comparison, and has an air flow split ratio of 0 (100% primary air and 0% secondary air), and same residence time split as the RQL setting. The mechanism used in the simulation is ‘SYN-Mech’. When combustor outlet temperature is above 1670K, NO_x emissions of RQL are significantly reduced compared with the base line.

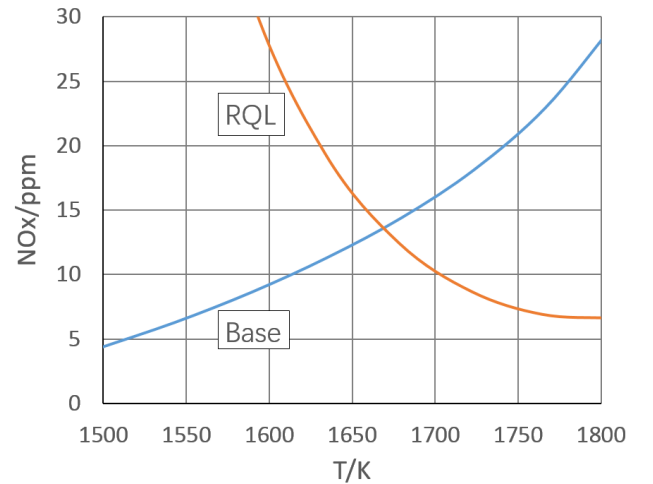


Figure 2 NO_x Emissions with Outlet Temperature

The RQL line decreases when the combustor outlet temperature increases. To explain the phenomena, NO_x produced in both rich-burn zone (9 PSRs) and lean-burn zone (the PFR) have been calculated (shown in Figure 3). The concentration of NO_x produced in PSRs is corrected to the PFR outlet condition. NO_x produced in PSRs is much higher than in PFR, and changes rapidly with the outlet temperature.

Meanwhile, NO_x produced in PFR changes little. Thus, NO_x produced in PSR zone (rich-burn zone) has major influence. Higher outlet temperature means larger equivalence ratio of the PSR zone, as shown in Figure 4. The relationship between NO_x and equivalence ratio in a single PSR is shown in Figure 5 (the pressure is 12atm, residence time is 5ms, and preheat temperatures of both fuel and air are 600K, the vertical axis is logarithmic). Under rich-burn condition, higher equivalence ratio means lower NO_x formation. Therefore, NO_x produced in rich-burn zone decrease with combustor outlet temperature. In addition, NO_x produced in PSR zone is dominating, as already shown in Figure 3. Thus, NO_x emissions in RQL (Figure 2) drop with temperature. And RQL combustor significantly reduces NO_x emission when combustor outlet temperature is higher than 1670K.

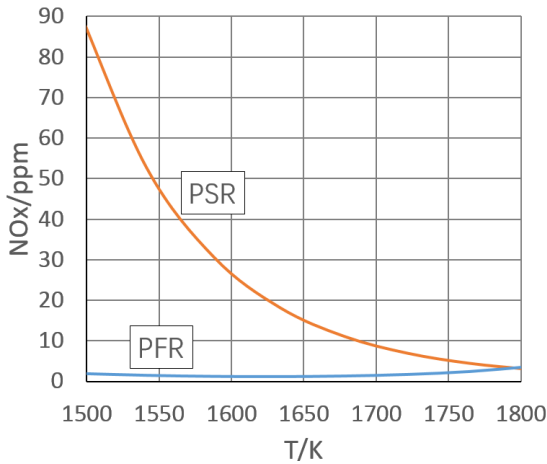


Figure 3 NO_x Produced in PSR and PFR

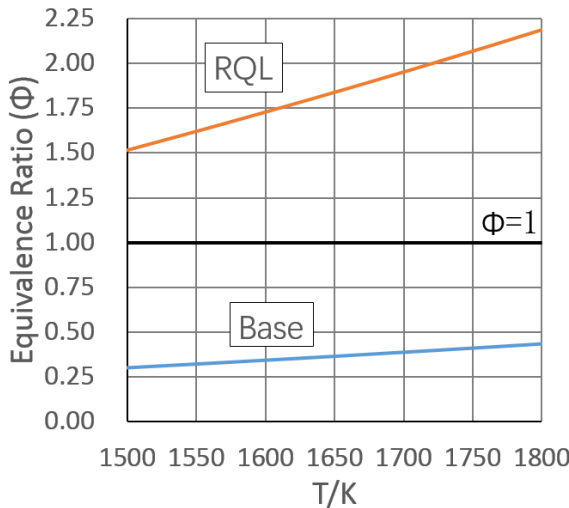


Figure 4 Equivalence Ratios of PSR Zone

Figure 6 presents the influence of air flow split on NO_x emissions of RQL combustor. The vertical axis of Figure 6 is logarithmic. The residence time of rich-burn zone is 5ms and that of lean-burn zone is 15ms. The outlet temperatures are 1750K and 1908K. The overall air flowrate and fuel flowrate are fixed under each temperature. All fuel is injected into the rich-burn area. Thus, higher air flow split ratio results in higher equivalence ratio in rich-burn area. When the rich-

burn zone is near the stoichiometric condition (air split ratio of 1.5), extremely high NO_x emissions appear. With higher air flow split ratio, the NO_x emissions decrease instantly, due to lower flame temperature in the rich-burn zone.

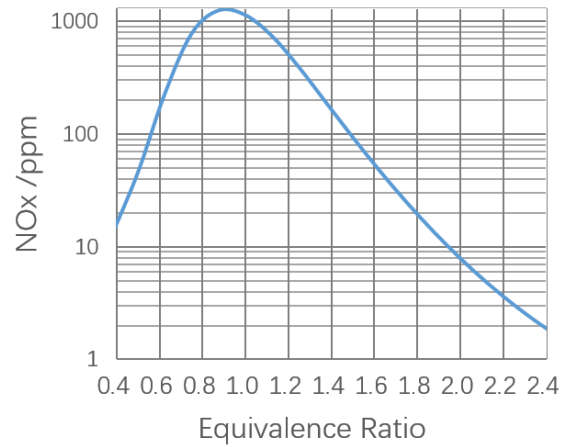


Figure 5 Influence of Φ on NO_x Formation in PSR

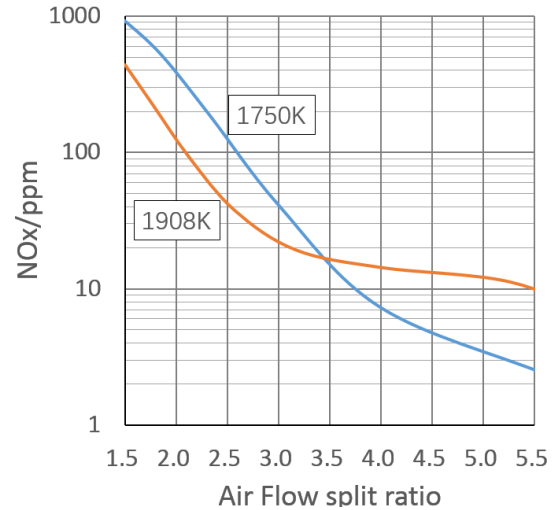


Figure 6 Influence of Air Flow Split

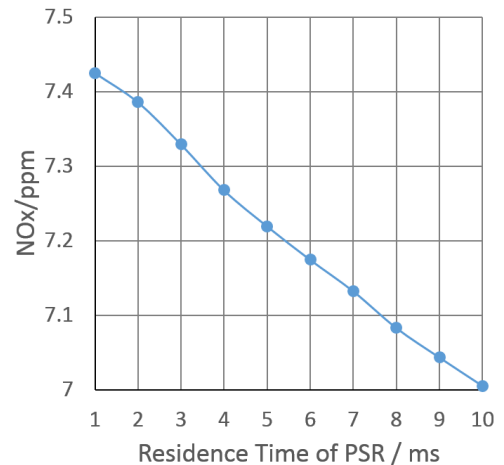


Figure 7 Influence of Residence Time Split

Figure 7 shows the influence of residence time split on NO_x emissions of RQL combustor. The total residence time of PSR and PFR zone is set to 20ms, and residence time of PSR zone are varied from 1ms to 10ms. The combustor

outlet temperature is 1750K. NO_x emissions decreases with longer residence time in PSR zone (less residence time in PFR zone). The decrease comes from the less thermal NO_x production in PFR due to less residence time. However, such decrease of NO_x is not significant compared with NO_x produced in PSR zone and produced at the beginning of PFR. Thus, very limit amount of NO_x can be reduced by increasing PSR residence time at this working condition.

Influences of mechanisms on NO_x prediction

GRI-Mech 3.0 mechanism is chosen for comparison. To investigate the differences, model with a single PSR and PFR was used, without considering the mixing effect in the rich-burn zone. Figure 8 shows the schematic of the model. Residence time of PSR is fixed to 5ms and 15ms in PFR. Air flow split is set to 20% primary air and 80% secondary air. Two cases are studied – outlet temperature at 1908K and 1750K.

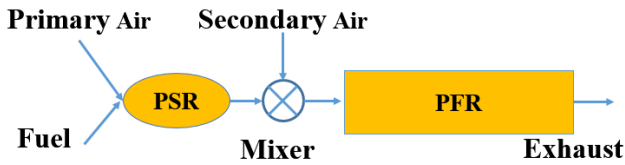


Figure 8 Schematic of Model for Mech. Study

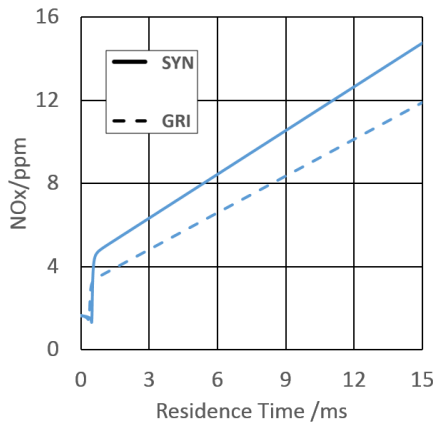


Figure 9 NO_x prediction in PFR

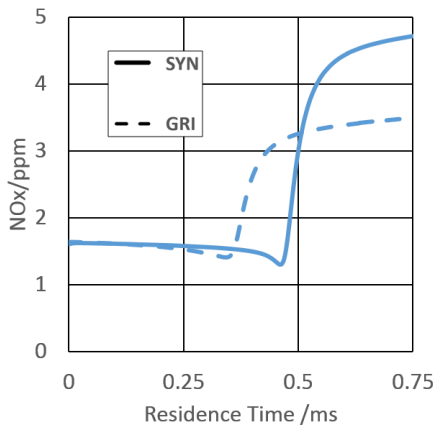


Figure 10 Closer look of NO_x concentration

Figure 9 shows NO_x concentration in the PFR, when the outlet temperature is 1908K. Mechanisms have almost the

same prediction of NO_x at the very begin of the PFR at 1.64ppm. While at PFR outlet (residence time of 15ms), the predictions are different. ‘SYN-Mech’ predicts 14.8ppm, while GRI-Mech 3.0 shows 11.9ppm.

Figure 10 gives a closer look of NO_x concentration at the beginning of the PFR. Both mechanisms predict a sharp increase of NO_x in less than 1ms and then a linear increase at different rates. ‘SYN-Mech’ predicts both the sudden increase and the linear-increase rate higher than GRI-Mech 3.0.

Temperature, major reactions that influence NO_x production and concentration of reactants are investigated to explain the differences between predictions of each mechanism. Since predictions at PFR inlet is almost same, the investigation is focused on the PFR. Figure 11 shows the change of temperature with residence time in PFR. Temperatures predicted by both mechanisms increase sharply at the beginning of PFR, and stay constant at 1908K after the initial time. Predictions of temperature are almost same of both mechanisms.

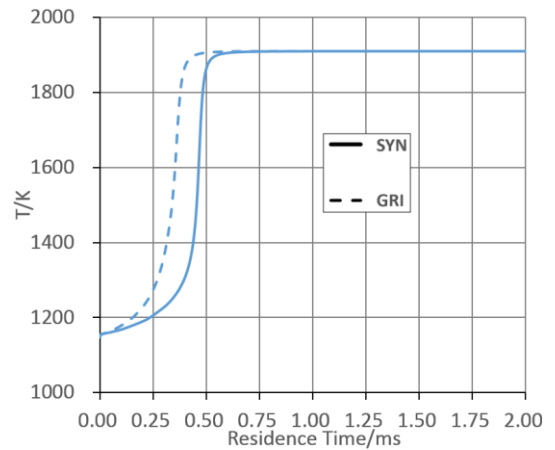
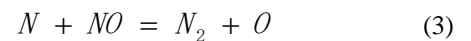


Figure 11 Temperature in PFR, 1908K Outlet

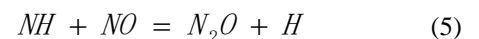
Considering the large amount of elementary reactions that these two mechanisms contains, major reactions that influence NO_x formation need to be found to enable further investigate on concentration of reactants. Sensitivity analyses were conducted to find these major reactions, using the CHEMKIN-PRO software package. Figure 12 shows NO sensitivity on pre-exponential factor “A” of the reaction rate constant of elementary reactions, and the most sensitive 4 of each mechanisms were selected on the figure.

These reactions happen to match three pathways of the NO formation (Correa, 1993) (Li et al. 2009) (Sahu et al. 2014).

Zeldovich (thermal) pathway:



N₂O pathway:



NNH pathway:

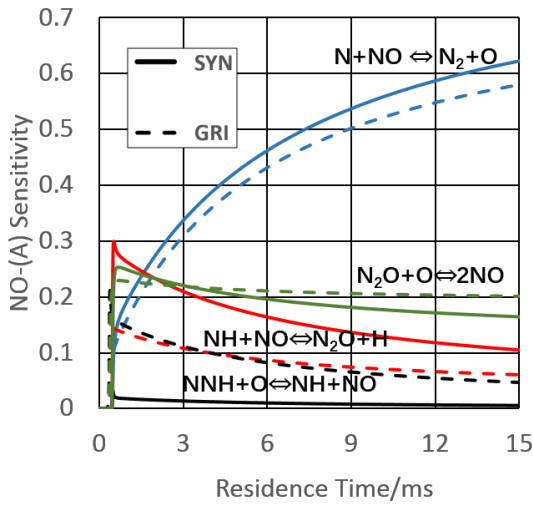
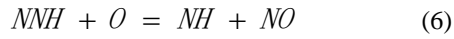


Figure 12 NO Sensitivity in PFR, 1908K

Figure 13 gives a closer look of NO sensitivity at the beginning of the PFR. NO sensitivities of all these pathways increase sharply, at the beginning. After the initial increase, both mechanisms predict that NO sensitivity of N_2O pathway and NNH pathway decrease, and NO sensitivity of thermal pathway increases. The most sensitive pathway initially is N_2O pathway in both mechanisms. NNH pathway is the least sensitive among major pathways in ‘SYN-Mech’, yet in GRI-Mech 3.0 it is much more sensitive at the beginning. NO sensitivity of NNH pathway and N_2O pathway reduce with residence time, while sensitivity of thermal pathway continually increases. Within 0.75ms, NNH pathway and N_2O pathway have the largest influence. When residence time increases, thermal pathway becomes the dominating one.

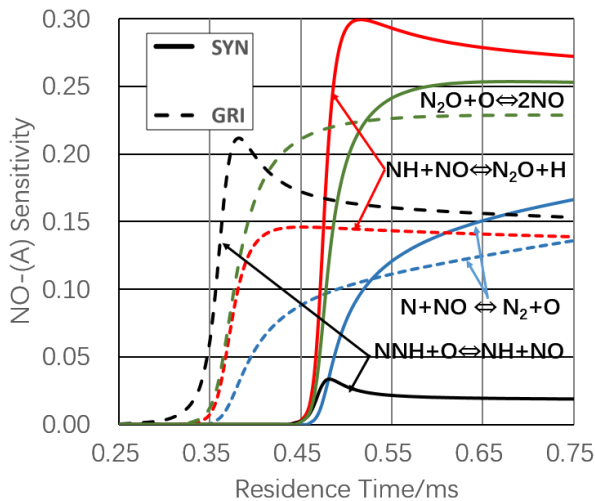


Figure 13 Closer Look of NO Sensitivity

In addition, some of the equations have NO as reactant because the reaction rate constant is larger than the reverse reaction rate constant. However, the positive sensitivity factor indicates that these pathways contribute to NO formation rather than reducing it.

Figure 14 and 15 shows the concentration of reactants of the major pathways in PFR. The results are NOT corrected to 15% O_2 and dry condition. The vertical axis of Figure 14 is logarithmic. All the reactant drops to low concentration after entering PFR, followed by a peak, then fall back to constant values. For O, N_2O and H, the constant values are same in both mechanism, and ‘SYN-Mech’ predicts higher peak (shown in Figure 14). For NNH, GRI-Mech predicts higher peak, and the constant value are not same, $8.3E-6$ ppm in ‘SYN-Mech’ and $1.3E-5$ ppm in GRI-Mech 3.0 (shown in Figure 15).

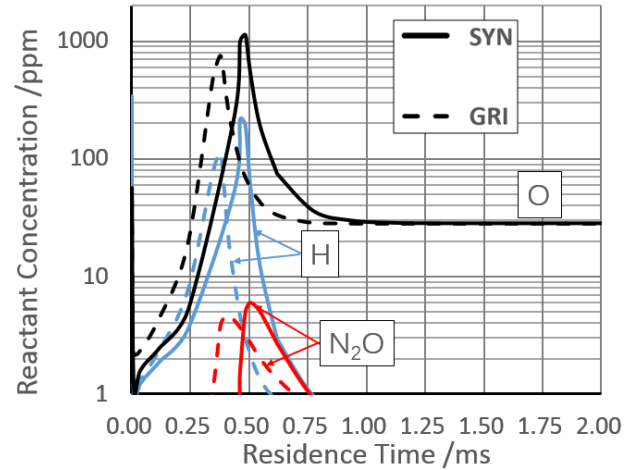


Figure 14 Concentration of Reactants, 1908K

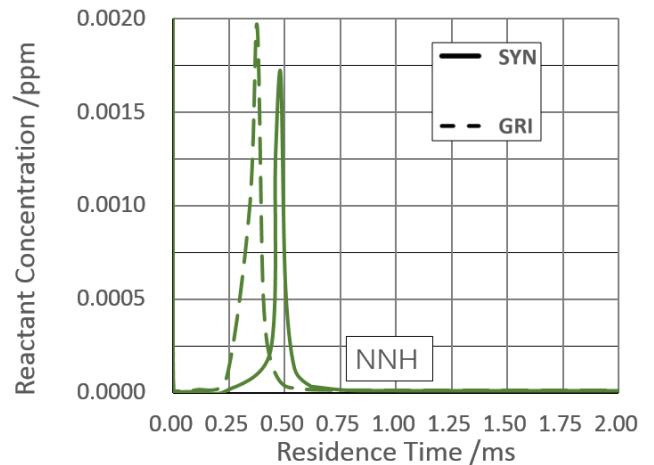


Figure 15 Concentration of NNH, 1908K

To further investigate the contribution to NO formation of each pathway, rate of production (ROP) has been calculated. Figure 16 and Figure 17 present ROP with residence time lower than 1ms, matching the temperature increasing range. In the sharp temperature-increase zone inside PFR, both mechanism consider N_2O pathway having the highest ROP of NO. NNH pathway is considered more important in GRI-Mech 3.0 than in ‘SYN-Mech’. ROP of N_2O pathway of ‘SYN-Mech’ is much higher than in GRI-Mech 3.0. Thus, ‘SYN-Mech’ predicts higher NO concentration at the beginning of PFR.

Figure 18 shows ROP after 1ms in PFR. ROP of each pathway is almost constant due to the constant temperature

and concentration of reactants. Thermal pathway dominates in both mechanisms, followed by N_2O pathway. ‘SYN-Mech’ predicts a higher ROP of thermal pathway, and a lower ROP of N_2O pathway, compared with GRI-Mech 3.0. The total ROP of these major pathways is higher in ‘SYN-Mech’. Therefore, ‘SYN-Mech’ predicts a higher increasing rate of NO when the temperature becomes constant.

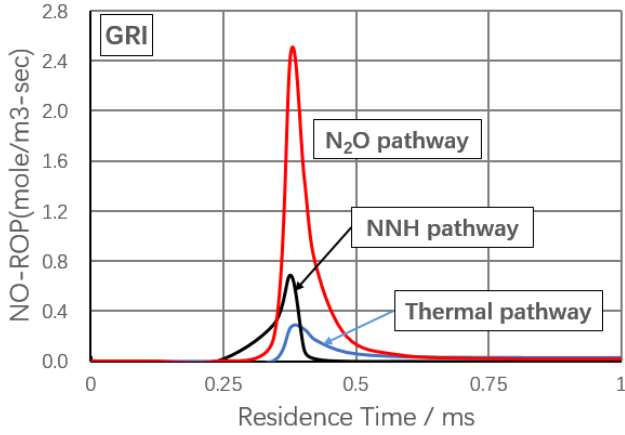


Figure 16 ROP within 1ms of GRI-Mech 3.0, 1908K

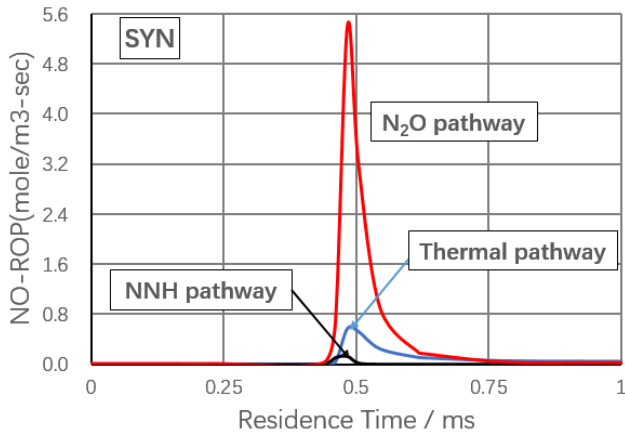


Figure 17 ROP within 1ms of ‘SYN-Mech’, 1908K.

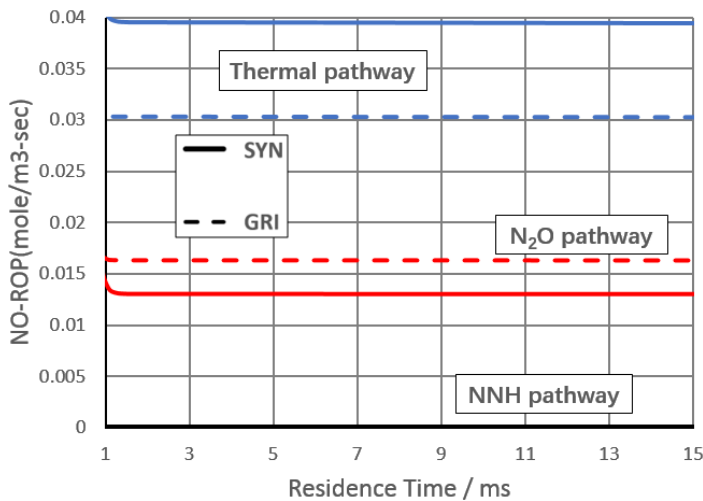


Figure 18 ROP after 1ms of in PFR, 1908K

Combining Figure 16 to 18, both higher prediction of NO within sharp temperature-increase zone and constant

temperature zone contribute to the higher NO_x prediction of ‘SYN-Mech’ when outlet temperature is 1908K.

The 1750K case is another story, in which mechanisms have different predictions in both PSR (PFR inlet) and PFR, as shown in Figure 19. Figure 19 a) is corrected to 15% O₂ and dry condition while Figure 19 b) is corrected to ONLY dry condition. ‘SYN-Mech’ predicts lower overall NO_x emission, different from the 1908K case. In in Figure 19 a), the NO_x drop at PFR beginning is caused by the 15% O₂ correction, as O₂ concentration changes rapidly when O₂ reacts with extra fuel from PSR zone at the beginning. Figure 19 b) shows that both mechanisms predict a sharp NO_x increase at the beginning of PFR followed by a linear increase. ‘SYN-Mech’ predicts both higher initial increase and linear-increase speed, but a lower NO_x concentration at PFR inlet. To explain these, sensitivity analyses have been conducted concerning both PSR and PFR, and major reactions contributed to NO production have been selected. ROP of these reactions are then calculated.

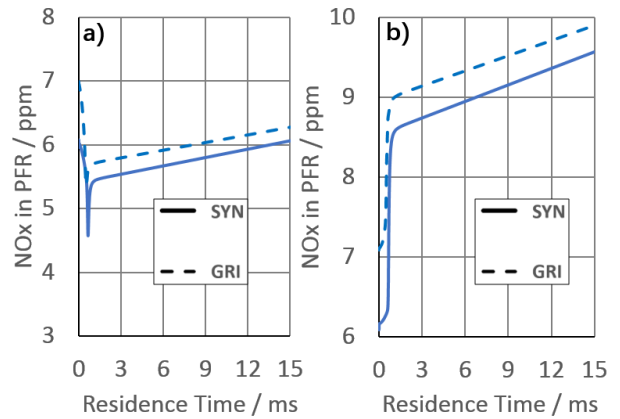


Figure 19 NO_x prediction in PFR, 1750K

Corrected to a) 15% O₂, Dry; b) Dry

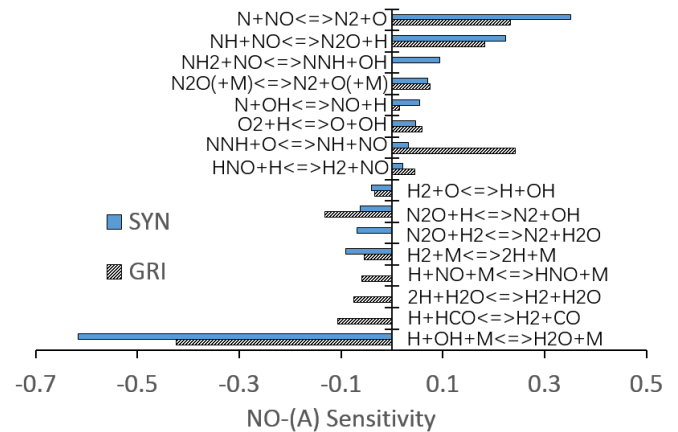


Figure 20 NO Sensitivity in PSR, 1750K

Figure 20 shows the most sensitive reactions in PSR. The major ones are thermal pathway, N_2O pathway and NNH pathway. In the prediction of ‘SYN-Mech’, thermal pathway dominates. However, in prediction of GRI-Mech 3.0, NNH pathway dominates. ROP of these major reactions is shown in Figure 21. Both mechanisms predict similar ROPs of

thermal pathway and N_2O pathway. But GRI-Mech 3.0 predicts a much higher ROP of NNH pathway. The total ROP predicted by GRI-Mech 3.0 is higher, leading to higher NO_x concentration in PSR.

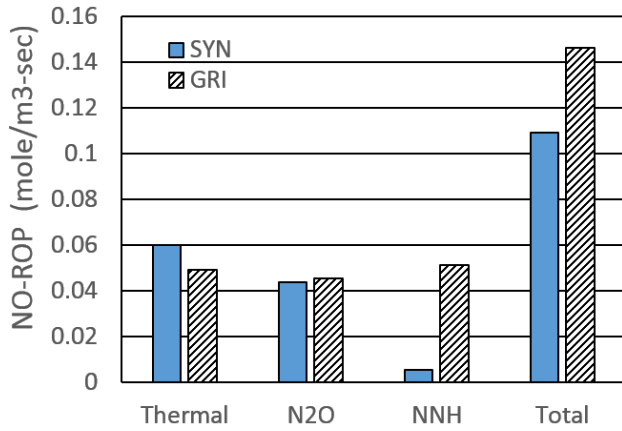


Figure 21 ROP in PSR, 1750K

Figure 22 shows the most sensitive reactions in PFR. The N_2O pathway is dominating, and ‘SYN-Mech’ predicts higher NO sensitivity on N_2O pathway, compared with GRI-Mech 3.0.

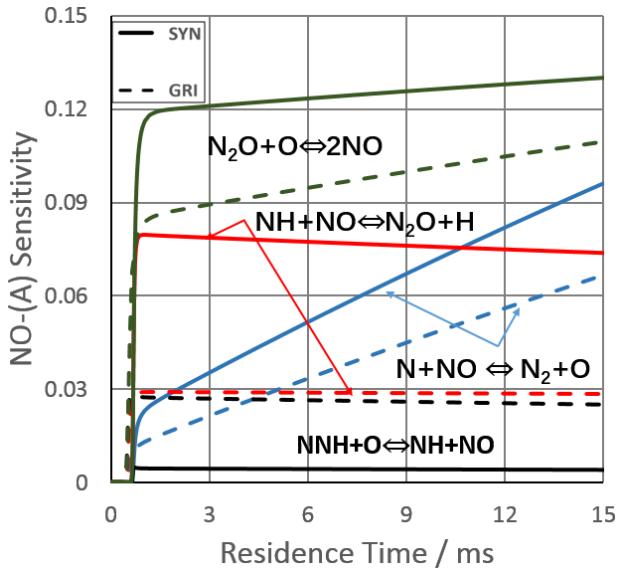


Figure 22 NO Sensitivity in PFR, 1750K

Figure 23 presents the ROP in PFR within 1ms, ‘SYN-Mech’ predicts much higher ROP peak of N_2O pathway (similar with the 1908K case) and NNH pathway (different from the 1908K case). Figure 24 presents the ROP in PFR after 1ms. ROPs of all pathways drop to a constant value, and ‘SYN-Mech’ predicts higher ROP of thermal pathway, lower ROP of N_2O pathway, and also predicts a higher total ROP. Thus, it predicts higher NO production in PFR, similar with the 1908K case.

In the 1750K case, ‘SYN-Mech’ considers a higher NO production within the PFR, compared with GRI-Mech 3.0. However, the lower NO prediction in PSR leads to the lower overall NO_x prediction.

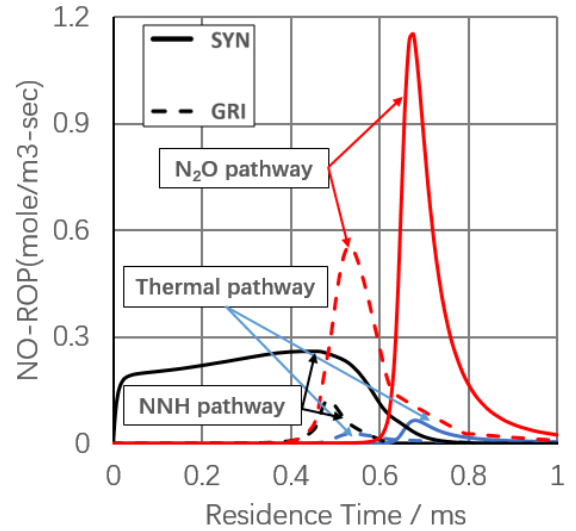


Figure 23 ROP within 1ms of in PFR, 1750K

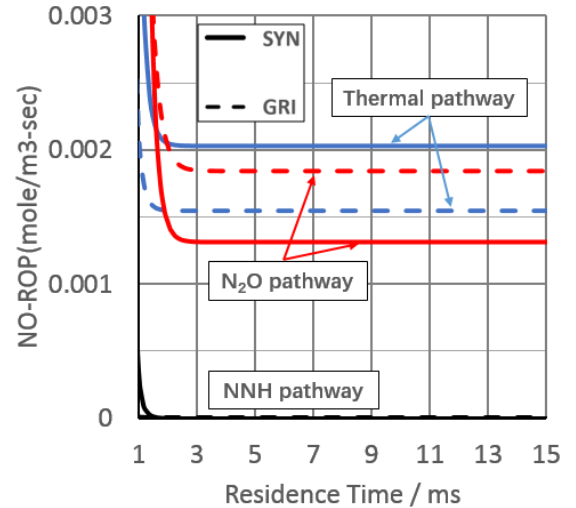


Figure 24 ROP after 1ms of in PFR, 1750K

CONCLUSIONS

Chemical reactor network (CRN) model is used to investigate NO_x emission in RQL combustor. With fixed air flow split, RQL combustor significantly reduces NO_x emissions at high temperature. Influences of air flow split and residence time split are also investigated, with fixed outlet temperature. The air flow split is the key parameter - NO_x emissions drop significantly when less primary air is injected to rich-burn zone.

Influences of reaction mechanisms on NO_x prediction are studied:

- (1) In the 1908K case, ‘SYN-Mech’ predicts 24% higher overall NO_x emissions compared with GRI-Mech 3.0. ‘SYN-Mech’ predicts similar NO_x production in PSR and higher NO_x production in PFR. In PFR, ‘SYN-Mech’ predicts higher ROP of N_2O pathway at the beginning and then higher ROP of thermal pathway after temperature reached the constant 1908K, compared with GRI-Mech 3.0.
- (2) In the 1750K case, ‘SYN-Mech’ predicts 4% lower overall NO_x emissions compared with GRI-Mech 3.0.

‘SYN-Mech’ predicts lower NO_x production in PSR and higher NO_x production in PFR. In PSR, both mechanisms predict similar ROPs of thermal and N₂O pathway. However, ‘SYN-Mech’ predicts much lower ROP of NNH pathway, leading to lower NO_x prediction in PSR. Situation in PFR is same with the 1908K case.

NOMENCLATURE

ϕ equivalence ratio
 σ standard deviation
 η mixing efficiency
 A pre-exponential factor of the reaction rate constant

ABBREVIATION

CRN chemical reactor network
 IGCC integrated gasification combined cycle
 PFR plug flow reactor
 PSR perfect stirred reactor
 ROP rate of production
 RQL rich burn quick mix lean burn

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