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Analysis of air-staged combustion of NH_3/CH_4 mixture with low NO_x emission at gas turbine conditions in model combustors

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ABSTRACT

The NO_x formation characteristics have been numerically investigated in the NH_3/CH_4 fired model combustor consisting of perfectly stirred reactors (PSR) and plug flow reactors (PFR) with 84-species Tian mechanism at gas-turbine relevant conditions of 23 atm and 1873 K at the combustor outlet. Specifically, the impacts of NH_3 dilution on NO_x formation were first studied in a non-staged model combustor consisting of one PSR representing main combustion zone and one PFR representing post-combustion zone over a wide range of the volumetric ratio of NH_3 in the NH_3/CH_4 fuel. It was found that the NO_x emission increases sharply with the NH_3 addition mainly through the enhanced HNO pathway in the main reaction zone. Recognizing that the NO_x formation via the HNO pathway is limited by the availability of oxygen, the effects of fuel/air equivalence ratio on NO_x formation were then quantified under different levels of NH_3 dilution. It was revealed that at fuel-rich conditions, the NO_x formation via HNO pathway can be offset by the reduction via other pathways, such as pathways related to NHi, leading to the overall low NO_x emission. Finally, an air-staged model combustion system was proposed, in which the equivalence ratio in the primary stage was chosen to be 1.5 to take advantage of the low NO_x emission under fuel-rich conditions. The air split among the two stages was determined by the fixed outlet temperature of 1873 K. A parametric study demonstrated that less than 30 ppm NO_x emission can be achieved with simple two-stage combustion even when the volumetric ratio of the NH_3 reaches 40%, which is dramatically lower than the one of thousands ppm in the non-staged combustion. The results were further verified with the 47-species Konnov mechanism, demonstrating the great potential of air-staged combustion for low NO_x emission in the NH_3/CH_4 fired combustion.

1. Introduction

To protect against global warming, NH_3 has been reconsidered as an alternative fuel in recent years due to its chemical features of high energy density and non-carbon carrier compared to traditional fossil fuels and the characteristics of easier and less expensive storage and delivery compared to H_2 [1–4]. The NH_3 fired combustion is not a novel concept and it has been explored for engine applications in early 1940s [5] with the remaining technical challenges of high minimum ignition energy (MIE), combustion instability and particularly high NO_x emission to be addressed [6].

The earlier development of an NH_3 gas turbine had been discarded because of the extremely low combustion efficiency and combustion instability [7]. Recent demand for H_2 energy carrier revives the usage of NH_3 fuel and significant progresses have been made toward an application of NH_3 gas turbine power generation. Kobayashi et al. [3,6,8–11] have performed systematic studies on combustion and emission characteristics of NH_3 fired combustion, which lead to successful application on NH_3-kerosene gas turbine power generation [6], and later pure NH_3 gas turbine power generation [3]. In the first successful NH_3 fired gas turbine application, NO_x emission less than 10 ppm was achieved in conjunction with a NO_x selective catalytic reduction (SCR) apparatus.

Recently, it is proposed to blend NH_3 with other energetic fuels such as CH_4 or H_2 for stable combustion although the tradeoff between stability and emission requires further studies. Some experimental and numerical studies have been performed to analyze the combustion characteristics of NH_3/CH_4 or NH_3/H_2 fuels [12–15]. Valera-Median et al. [12,13] showed that stable flames of NH_3/CH_4/air can be achieved with low NO_x emission in a tangential swirl burner, but in a narrow operational range with the equivalence greater than one. The emission characteristics together with autoignition and flame

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propagation properties of NH_3/CH_4 under gas turbine conditions have been numerically studied by Xiao et al. [14,15] using idealized homogeneous reactors and freely propagating laminar flames with different chemical mechanisms [16,17]. It was shown that with an increased amount of NH_3, the ignition delay time of the fuel/air mixtures increases while the flame speed decreases. More importantly, the equivalence ratio plays a key role for the NO_x emission and richer NH_3 condition was suggested to ensure lower NO_x emission through the recombination of NH_3 with NO_x [15]. Lee et al. concluded that the potential of significant NO_x reduction due to the substantially reduced temperature with ammonia substitution may be overweighed by the enhanced NO_x formation effect induced by the NH_3-containing N through the experiments carried out in a spherical windowed chamber [18]. Li et al. experimentally studied the NO_x formation characteristics for NH_3/H_2 fuel mixture at different equivalence ratios in a stainless tube-type burner under atmospheric pressure [19]. It was illustrated that NH_3 can be consumed completely even with an excess of fuel due to the reaction with NO_x, but would result in excessive H_2 similar to NH_3/CH_4 fuel system. Duynslaegher et al. [20–22] discussed the flame structure and emission characteristics of premixed NH_3 fired combustion at different pressures and equivalence ratios. It was observed that a decrease of the equivalence ratio results in an increase of the NO_x formation.

As a N-containing fuel, one challenge for the use of ammonia in combustion systems is the high production of fuel NO_x [23,24]. Although the fuel-rich condition in NH_3 fired combustion is confirmed above to be advantageous for low NO_x formation, it is accompanied with incomplete combustion, while for lean combustion the NO_x emission would in general not meet the requirements of NO_x emission without proper combustion organization or after-treatment. The SCR or non-selective catalytic reduction (NSCR) methods [9], though effective for NO_x reduction, add significant infrastructure and operation costs. More recently, several advanced low-NO_x combustion techniques for NH_3 fired systems such as staged combustion have been analyzed through both experimental and numerical studies. Staged combustion with NH_3 in fuel has been investigated in [25] with the equivalence ratio being 1.6 in the fuel-rich stage and the results show that staged combustion is promising for NO_x reduction under gas turbine conditions. Although combustion technologies for NO_x reduction such as staged combustion and nitrogen dilution were discussed in [25], the work was mainly focused on the analysis of the performance of gasified fuels utilized in integrated gasification combined cycle (IGCC) in which the NH_3 was not removed thoroughly in the nonpremixed combustion. The detailed characteristics of NO_x emission in the NH_3 fired combustion are still not clear in the main combustion zone and post-combustion zone and there are still very few studies on analyzing the use of staged combustion for NO_x reduction at the operating conditions of next generation gas turbine. It is therefore of practical importance to analyze the emission characteristics in NH_3-fired combustion systems at engine relevant conditions and explore possible staged combustion technique for low NO_x emission.

The study contributes to low NO_x emission for NH_3/CH_4/air gas turbine combustion through a systematic study of NO_x formation mechanisms over a wide range of equivalence ratio and NH_3 dilution ratio. Through proposing, analyzing and optimizing the air-staged model combustion system for lean NH_3/CH_4/air gas turbine combustion, it demonstrates that less than 30 ppm NO_x emission can be achieved at gas turbine conditions even when the volumetric ratio of the NH_3 reaches 40%. The remainder of the paper is organized as follows. In Section 2, the effects of NH_3 dilution on NO_x formation were first studied in a non-staged NH_3/CH_4 fired model combustor, with the contributions from different NO_x reaction pathways being quantified. Then, in Section 3, an air-staged model combustion system was proposed for low NO_x emission in NH_3/CH_4/air fired combustions, with its emission characteristics being thoroughly analyzed. Conclusions are in Section 4.

Table 1
Pressure and temperature parameters.

Fuel	p (atm)	T_{unburn} (K)	T_{burn} (K)
NH_3/CH_4	23	647	1873

2. Emission characteristics of premixed NH_3/CH_4 fired combustion

The emission characteristics are analyzed based on the operating conditions of state-of-the-art H/J class heavy duty gas turbines, in which the temperature at the outlet of the combustor or at the first-stage rotor blade ranges from 1773 to 1873 K and the pressure is larger than 20 atm in the combustor. The specific operating conditions are listed in Table 1 as in [26]. In this work, the approach of chemical reactor network [27] is employed to analyze the emission characteristics of premixed NH_3/CH_4 fired combustion. For example, the non-staged model combustor consists of a PSR representing the main combustion zone and a PFR representing the post-combustion zone. The residence times in the PSR and the PFR are specified according to the representative residence times in real combustion devices [28], which are around several milliseconds in the main combustion zone and around 20 ms in the post-combustion zone for heavy duty gas turbines.

For the idealized zero-dimensional homogenous PSR and one-dimensional PFR, the evolution of species and energy is governed by

$$\mathrm{PSR}{:}\rho\frac{Y_k^{in}-Y_k}{\tau_{PSR}} = -W_k\dot{\omega}_k \tag{1a}$$

$$\rho\frac{\sum_{k=1}^{Ns}(Y_k^{in}h_k^{in}-Y_kh_k)}{\tau_{\mathrm{PSR}}} = -\sum_{k=1}^{Ns}h_kW_k\dot{\omega}_k \tag{1b}$$

$$\mathrm{PFR}{:}\rho\frac{dY_k}{dt} = W_k\dot{\omega}_k \tag{2a}$$

$$\rho c_p\frac{dT}{dt} = -\sum_{k=1}^{Ns}h_kW_k\dot{\omega}_k \tag{2b}$$

in which Y_k, W_k, h_k, $\dot{\omega}_k$ are the mass fraction, molecular weight, enthalpy and net reaction rate of the k^{th} species, respectively, ρ is mixture density, c_p is specific heat, τ_{PSR} is the residence time in the PSR, Ns is the number of species, and the superscript in represents inlet. Note that the coordinate transformation has been performed for PFR to convert from spatial to time coordinate. In this study, the modules of PSR and PFR in CHEMKIN [29] are employed for calculations. The Tian mechanism [16] with 84 species and 703 reactions is employed, which has been applied in several NH_3 fired combustion applications demonstrating good performance [8,23]. The reduced Konnov mechanism with 47 species and 500 reactions [15] is also employed to investigate the dependence of emission characteristics on chemical mechanisms.

The mole fraction of NH_3 in the NH_3/CH_4 mixture is denoted as η and the stoichiometric global reaction of NH_3/CH_4/air is given by

$$\eta NH_3 + (1-\eta)CH_4 + \left(2-\frac{5}{4}\eta\right)(O_2 + 3.76N_2) = (1-\eta)CO_2 + \left(2-\frac{1}{2}\eta\right)H_2O + (7.52-4.2\eta)N_2 \tag{3}$$

With a given amount of NH_3 dilution η, to maintain the specific engine operation temperature of 1873 K, an iterative procedure at a fixed pressure is employed to determine the unburnt mixture i.e., the equivalence ratio. The amount of fuel is $\phi[\eta NH_3 + (1-\eta)CH_4]$ when the system is not at stoichiometric. Fig. 1 shows the variation of the unburnt mixture against NH_3 dilution. As expected, both CH_4 and O_2 decrease with the increased NH_3 dilution. In addition, the equivalence ratio increases slightly with NH_3 dilution to offset the low energy density of NH_3, but maintains fuel-lean condition even with $\eta = 100\%$. In the following, the emission characteristics are analyzed in the main