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A Numerical Study on the Effect of Water Addition on NO Formation in Counterflow CH_4/Air Premixed Flames

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The effect of water addition on NO formation in counterflow CH_4/air premixed flames was investigated by numerical simulation. Detailed chemistry and complex thermal and transport properties were employed. The results show that the addition of water to a flame suppresses the formation of NO primarily due to flame temperature drop. Among a lean, a stoichiometric, and a rich premixed flame, the effectiveness of water addition is most significant for the stoichiometric flame and least for the rich flame. The addition of water also reduces the formation of NO in a flame because of the chemical effect. Compared to the stoichiometric flame, the chemical effect is intensified in the lean and rich flames. [DOI: 10.1115/1.2432890]

Keywords: premixed flame, water addition, numerical simulation, NO_x

1 Introduction

Li and Williams [1] indicated that the addition of water suppresses NO_x formation through both the thermal and chemical effects in two-stage methane/air flames. Zhao et al. [2], Park et al. [3], and Hwang et al. [4] showed that although the concentration of OH is increased, the formation of NO is decreased, when steam is added to a diffusion flame. Bhargava et al. [5] illustrated that the addition of moisture to air stream also brings a significant reduction in NO_x emission in premixed flames. However, only lean premixed flames were studied, and a perfectly stirred reactor network code was employed for the simulation. Therefore, it is of interest to further investigate the effect of water addition on NO_x formation in premixed flames over a wide equivalence ratio range by a more detailed flame model.

The purpose of this technical brief is to numerically investigate the effect of water addition on the formation of NO in counterflow CH_4/air premixed flames, with equivalence ratio varying from lean to rich.

2 Numerical Model and Result Discussion

The flame configuration studied was an axisymmetric laminar counterflow premixed flame [6]. The calculation method can be found elsewhere [7]. The chemical reaction mechanism used was GRI-Mech 3.0 [8]. The pressure and the fresh mixture temperature were, respectively, 1 atm and 300 K. The simulations were conducted for three typical premixed flames: a stoichiometric flame ($\phi=1.0$), a lean flame ($\phi=0.7$), and a rich ($\phi=1.3$) flame. Stretch rate was 45 s^{-1} for all the flames studied.

The fraction of added water changed from 0% to 20%. The definition of the water fraction is $\alpha_{H2O}=V_{H2O}/(V_{H2O}+V_{CH4})$, with V_{H2O} and V_{CH4} being, respectively, the volume flow rates of water and fuel. The added water was assumed to be steam with the same temperature as CH_4/air mixture. In the presentation of results, NO emission index (EI_{NO}) is defined as the ratio of the formed NO to the consumed fuel, and the normalized NO emission index (β_{NO}) is the ratio of NO emission index from a flame with water addition to that from a flame without water addition.

Figure 1 shows the drop of the maximum temperature, when water is added. It is illustrated that the temperature drop is greatest for the rich flame and smallest for the lean flame. The flame temperature drop can cause the reduction of NO formation in a flame. We call this thermal effect. In addition, there may be some variation in chemical reaction rates owing to the addition of water, which also changes the formation of NO in a flame. This is referred to as the chemical effect.

To identify the relative importance of the thermal and chemical effects, an extra calculation was carried out for each flame. In this extra calculation, the added water was replaced by an artificial inert species (XH_2O). The artificial species has the same thermal and transport properties as water, but does not participate in any chemical reaction. Therefore, the addition of XH_2O suppresses NO formation only through the thermal effect. As a result, the difference between the results from the normal and the extra simulations is caused by the chemical effect. The disparity between the results from the flame without water or XH_2O addition and the flame with XH_2O addition is attributed to the thermal effect.

Figure 2 illustrates the effect of water addition on the normalized NO emission indices of the three studied flames. It shows that although the addition of water suppresses NO formation in all the flames, the effectiveness varies. It is most efficient for the stoichiometric flame ($\phi=1.0$) and least efficient for the rich flame ($\phi=1.3$). This differs from the flame temperature variation, as in Fig. 1. In addition, Fig. 2 also reveals that the addition of water causes the reduction of NO in a premixed flame through both the thermal and chemical effects. Although the chemical effect is much smaller than the thermal effect for all the three flames, it is relatively enhanced in the lean and rich flames.

Figure 3 illustrates the integrated NO formation and destruction rates from the most significant reactions. The number of each reaction is based on the order of it in GRI-Mech 3.0 [8]. Among these reactions, R186, R187, and R189 are the three related to NO_2. The net contribution of these three reactions to NO formation is negligible. For rest of the reactions, the dominant ones for the three flames vary. Reactions R178 ($N+NO=N_2+O$) and R180 ($N+OH=NO+H$) are the two most significant for the stoichiometric flame, while the dominant ones become R179 ($N+O_2=NO+O$), R208 ($NNH+O=NH+NO$), and R214 ($HNO+H=H_2+NO$) for the lean flame. For the rich flame, the most important reactions are R180 and R214.

NO can be formed by four routes, i.e., the thermal, the prompt, the N_2O, and the NNH intermediate routes. It is clear that R178 and R208 belong to the thermal and NNH intermediate routes [7,9], respectively. Although R179 and R180 were defined as reactions belonging to the thermal route in many references, it should be noted that the atomic nitrogen participating in these two reactions might come from other routes. Reaction R214 is caused by HNO that may come from the prompt route and the NNH or N_2O intermediate route. Therefore, to figure out which route

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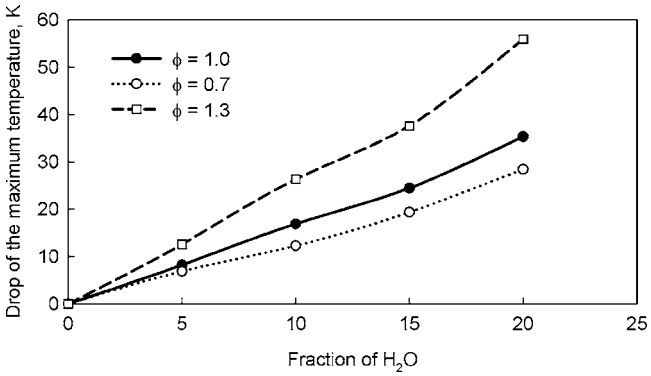

Fig. 1 Variation of the maximum flame temperature drop

dominates NO formation in a flame, we should further examine the rate of molecular nitrogen conversion to atomic nitrogen and species containing nitrogen element.

Figure 4 displays the integrated consumption rate of molecular nitrogen by different routes in the three flames. It is observed that in the stoichiometric flame, the thermal route dominates the conversion of molecular nitrogen. Although the thermal and prompt routes also contribute to the conversion of molecular nitrogen, the NNH and N_2O intermediate routes do the most in the lean flame. In the rich flame, almost all the molecular nitrogen conversion is through the prompt route. A pathway analysis shows that the most significant molecular nitrogen consumption reaction in the prompt route is R240 ($CH+N_2=HCN+N$). Therefore, we conclude that the thermal route dominates NO formation in the stoichiometric flame, while the NNH and N_2O intermediate routes contribute the most NO in the lean premixed flame. In the rich flame, NO is mainly formed by the prompt route.

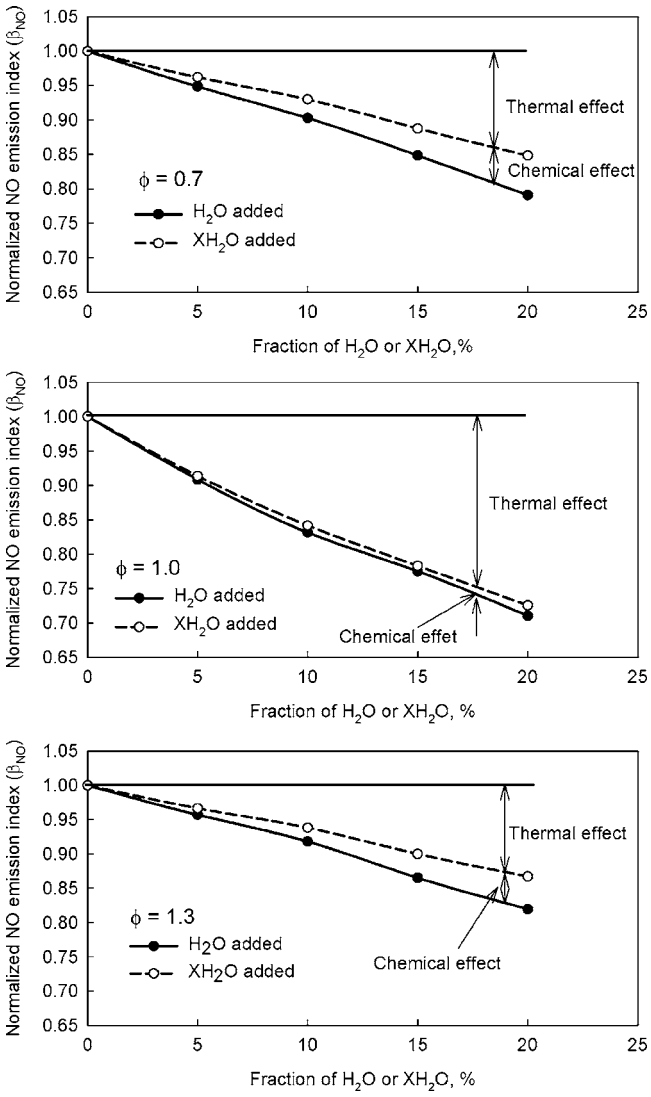

Fig. 2 Variation of normalized NO emission index

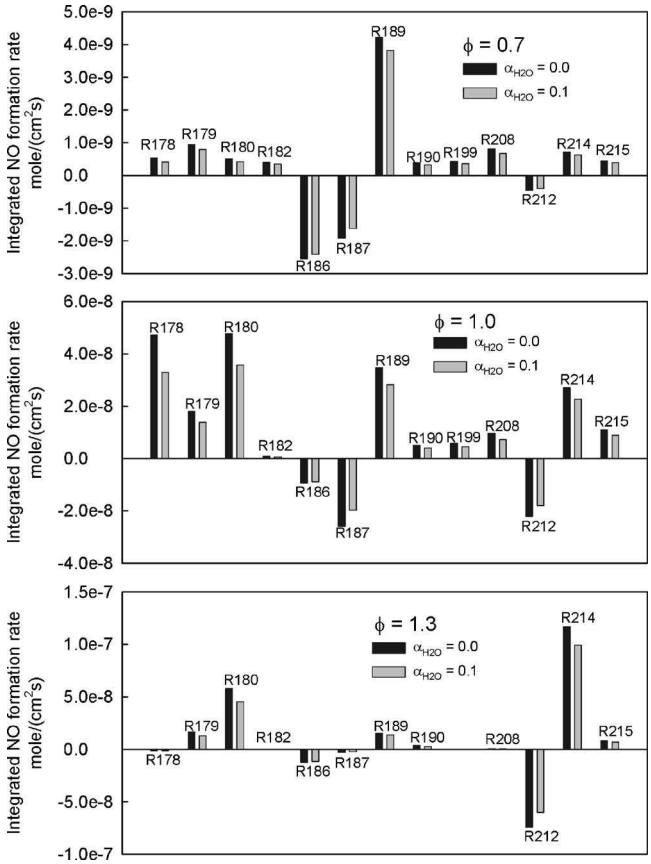

Fig. 3 Total NO formation and destruction rates

Since the thermal route forms NO in the high-temperature post-flame region, it is most sensitive to the variation in flame temperature. On the other hand, the prompt route usually forms NO in a very narrow low-temperature layer of flame front. Therefore, it is less sensitive to the variation of temperature. The NNH and N_2O intermediate routes dominate the formation of NO in the lean flame, but the thermal route also contributes some. Consequently, the effect of water addition on NO formation is most significant for the stoichiometric flame (mainly due to the thermal effect), and least for the rich flame, although the temperature drop in the rich flame is the greatest.

Now we examine how the addition of water causes the chemical effect on NO formation by comparing the results from the normal (H_2O addition) and the extra (XH_2O addition) simulations. The calculations first indicate that no recognizable difference can be observed in the maximum flame temperature for a flame between the results from the normal and extra simulations. However, the chemical effect of water addition does modify the concentrations of radicals OH, H, and O that affect the formation of NO, as shown in Fig. 5. For each flame, the maximum OH mole fraction from the simulation with H_2O addition is higher than that from the simulation with XH_2O addition, implying that the chemical effect

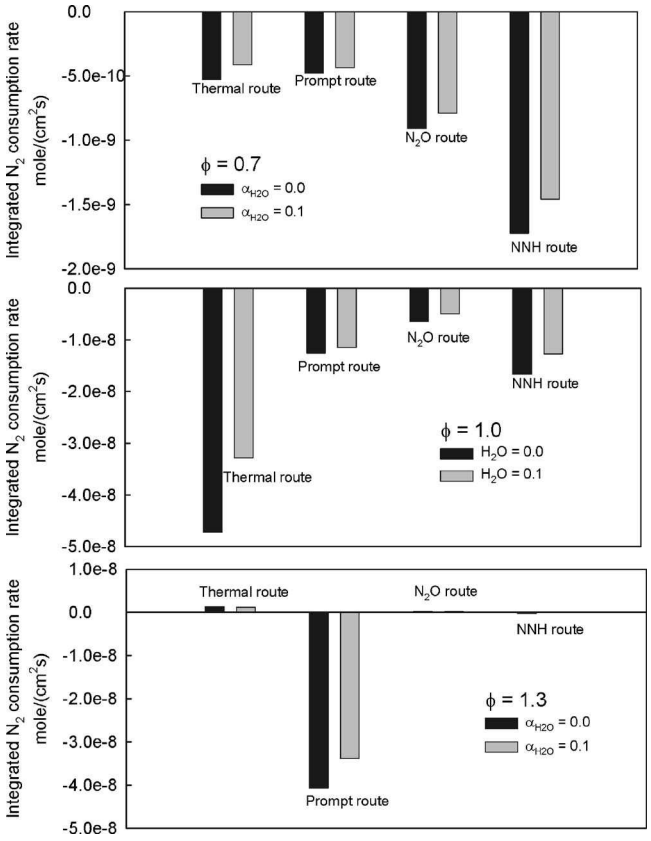

Fig. 4 Total molecular nitrogen consumption rate from different routes

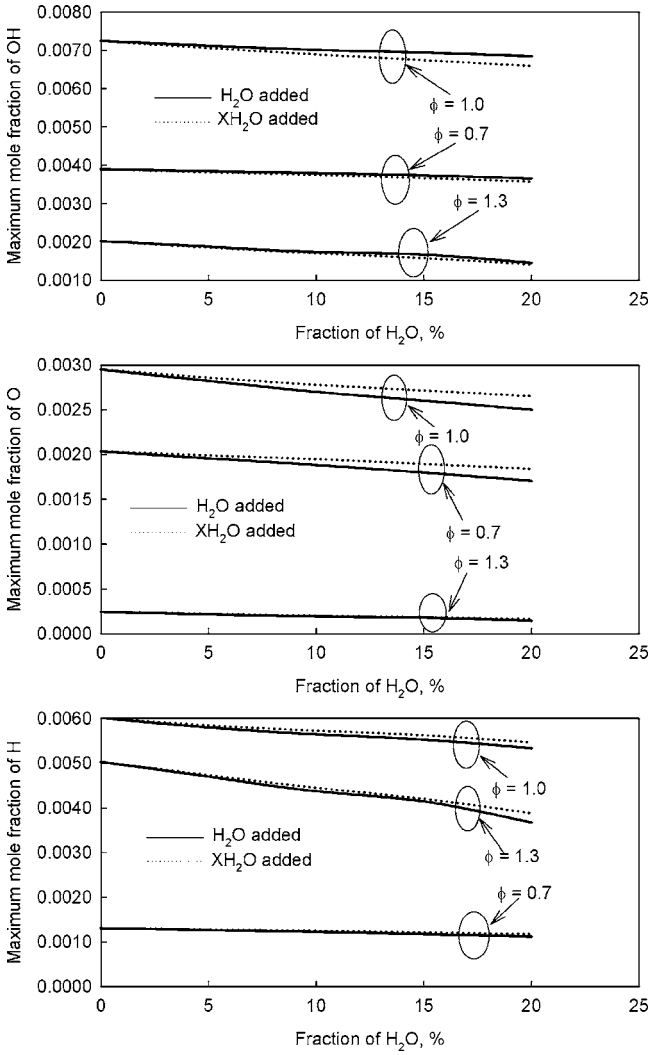

Fig. 5 Chemical effect of water addition on radicals OH, O, and H

of water addition actually enhances the production of OH. Reversely, the chemical effect of water addition reduces the concentrations of radicals O and H.

The formation of NO by the thermal route is through R178, R179, and R180. The chemical effect of water addition leads to negligible variation in flame temperature and increased OH concentration but decreased O concentration. Therefore, it has a very small net effect on NO formation from the thermal route, leading us to conclude that the chemical effect of water addition on NO formation in the stoichiometric flame is very small.

In the NNH and N_2O intermediate routes, the molecular nitrogen is first converted to NNH and N_2O. The formed NNH and N_2O are subsequently converted to NO. H and O radicals play significant roles in these conversion processes. Therefore, the chemical effect of water addition in the NNH and N_2O intermediate routes is bigger than in the thermal route, which leads us to conclude that the chemical effect of water addition in the lean flame is enhanced.

Molecular nitrogen is first converted to N atom and species containing nitrogen elements mainly by R240 in the prompt route. Then the N atom and HCN are converted to NO. The conversion rates of these reaction sequences are also closely related to O and H radicals. Besides, the chemical effect of water addition reduces the concentration of CH radical, as shown in Fig. 6 for the rich flame, which suppresses the initial conversion of molecular nitrogen by R240. As a result, the chemical effect of water addition on NO formation in the rich flame is also relatively bigger than in the stoichiometric flame.

3 Conclusions

This paper reports a numerical study on NO formation in counterflow CH_4/air premixed flames. The main conclusions are: (1) The addition of water to a flame suppresses the formation of NO primarily due to temperature drop. (2) The effectiveness of water addition for NO suppression is most significant for the stoichiometric flame and least for the rich flame. (3) The addition of water also reduces the formation of NO in a flame due to the chemical effect. (4) Compared to in the stoichiometric flame, the chemical effect is intensified in the lean and rich flames.

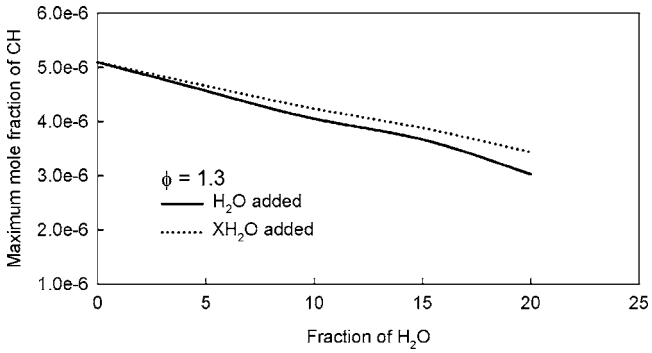

Fig. 6 Chemical effect of water addition on radical CH in the rich flame

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