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A Numerical Study on the Effect of CO Addition on Flame Temperature and NO Formation in Counterflow CH_4/Air Diffusion Flames

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A numerical study was carried out to understand the effect of CO enrichment on flame temperature and NO formation in counterflow CH_4/air diffusion flames. The results indicate that when CO is added to the fuel, both flame temperature and NO formation rate are changed due to the variations in adiabatic flame temperature, fuel Lewis number, and chemical reaction. At a low strain rate, the addition of carbon monoxide causes a monotonic decrease in flame temperature and peak NO concentration. However, NO emission index first slightly increases, and then decreases. At a moderate strain rate, the addition of CO has negligible effect on flame temperature and leads to a slight increase in both peak NO concentration and NO emission index, until the fraction of carbon monoxide reaches about 0.7. Then, with a further increase in the fraction of added carbon monoxide, all three quantities quickly decrease. At a high strain rate, the addition of carbon monoxide causes increase in flame temperature and NO formation rate, until a critical carbon monoxide fraction is reached. After the critical fraction, the further addition of carbon monoxide leads to decrease in both flame temperature and NO formation rate. [DOI: 10.1115/1.2906222]

Keywords: diffusion flame, NO_x, fuel enrichment, carbon monoxide

1 Introduction

Fuel enrichment is a promising concept for reducing fuel consumption and pollutant emission from combustion systems. Usually, hydrogen is selected as the additive for fuel enrichment combustion. Many studies have been conducted for some fundamental concepts of fuel enrichment combustion [1–4].

However, not enough attention has been paid to the effect of fuel enrichment on NO_x formation in diffusion flames. Naha and Aggarwal [5] investigated the effect of hydrogen addition on NO_x formation in strained nonpremixed methane and *n*-heptane flames at a fixed strain rate (100 s^{-1}). It was found that the addition of hydrogen has a minor effect on NO_x formation in methane (CH_4) flames and reduces the formation of NO_x in *n*-heptane flames. Our previous study [6] on the effect of hydrogen enrichment on NO formation in CH_4/air diffusion flames at various strain rates showed that the addition of a small amount of hydrogen has negligible effect on NO formation at low to moderate strain rates, but significantly increases NO formation at a high strain rate.

Although hydrogen has been shown to be an effective additive for fuel enrichment technology, it is only an energy carrier. It has to be generated from other fuels or water. A widely used method to obtain hydrogen is reforming hydrocarbon fuels. However, the reformate gas contains not only hydrogen but also carbon monoxide (CO) and some other minor components. If CO and other components contained in reformate gas do not have any negative side effects, or even helps in terms of improving combustion efficiency and reducing pollutant emission, we can directly use reformate gas as the additive in fuel enrichment combustion technology. Therefore, it is of interest to understand the effect of CO enrichment on combustion performance and pollutant emission.

In this paper, a detailed numerical study on the effect of CO addition on the formation of NO in CH_4/air diffusion flames with various strain rates was conducted. The fraction of CO changed from 0 to 0.9. The investigated strain rate covered a wide range.

2 Numerical Model

As in our previous investigation [6], an axisymmetric laminar counterflow flame configuration was employed, with fuel stream issuing from one nozzle and air from another. The simulations assumed the stagnation point flow approximation. The governing equations can be found elsewhere [7]. The calculations were carried out with a code revised from that of Kee et al. [8]. Upwind and center difference schemes were, respectively, used for the convective and diffusion terms in all the governing equations. Adaptive refinement of meshes was done to obtain grid independent results. Radiation heat loss was accounted for by an optically thin model [1].

The potential boundary conditions were used for freestream. The chemical reaction mechanism used is GRI-MECH 3.0 [9]. The thermal and transport properties were obtained by using the database of GRI-MECH 3.0 and the algorithms given in Refs. [10,11]. The pressure and the fresh mixture temperature were 1 atm and 298 K, respectively.

3 Results and Discussion

In all the studied flames, the fuel stream consists of CH_4 and CO. The fraction of CO is defined as $\alpha_{CO}=V_{CO}/(V_{CO}+V_{CH_4})$, with V_{CO} and V_{CH_4} being, respectively, the volume flow rates of CO and CH_4. The fraction of CO in this study covers a range from 0.0 to 0.9 for completeness.

3.1 Flame Temperature. Figure 1 shows the variation of peak flame temperature as the fraction of CO in fuel stream increases at three typical strain rates, a low ($a=10\ s^{-1}$), a moderate ($a=100\ s^{-1}$), and a high ($a=300\ s^{-1}$) one. Since the adiabatic equilibrium flame temperature (called adiabatic temperature hereafter) of CO is higher than that of CH_4 at stoichiometric condition, it was expected that the addition of CO to CH_4 would increase the peak flame temperature. However, the simulation results in Fig. 1 do not support this. Moreover, it is noted that the variation trend of peak flame temperature differs when strain rate is changed.

At $a=10\ s^{-1}$, the addition of CO causes a monotonic decrease in peak flame temperature. At $a=100\ s^{-1}$ or $300\ s^{-1}$, the peak temperature is almost constant or slightly increases and then decreases, as α_{CO} increases. These phenomena are due to the combined effects of adiabatic temperature, fuel Lewis number, chemical reaction, and residence time.

If the Lewis number is defined as the ratio of thermal diffusion rate to mass diffusion rate, the peak flame temperature of a diffusion flame will be increased or decreased when the Lewis number is less or greater than unity. The Lewis number of fuel is slightly

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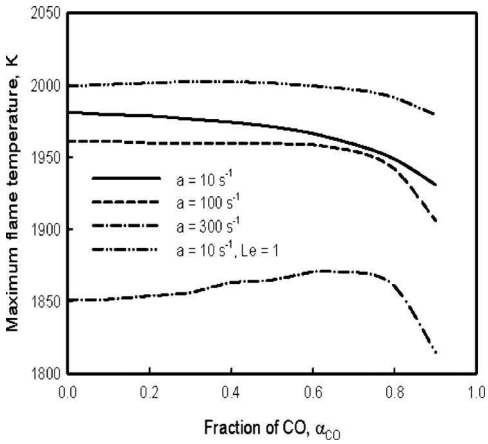

Fig. 1 Variation of peak flame temperature

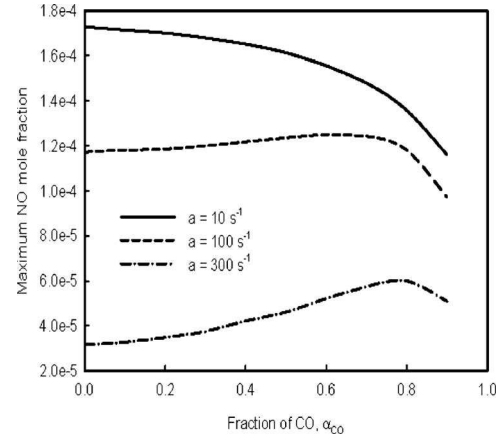

Fig. 2 Variation of peak NO mole fraction

less than unity for pure CH_4 diffusion flame. When CO is added, the fuel becomes a mixture. The Lewis number of CO is slightly greater than unity, leading to the slightly greater fuel Lewis number in CO enriched mixture than in pure CH_4. The increase in fuel Lewis number tends to decrease the peak flame temperature, which is one reason for the decrease in peak flame temperature at $a=10\ s^{-1}$ when α_{CO} is small. The almost constant and slight increase in maximum flame temperature at $a=100\ s^{-1}$ and $300\ s^{-1}$ will be explained later.

To confirm the effect of the fuel Lewis number, extra calculations were carried out at a strain rate of $10\ s^{-1}$ with the Lewis numbers artificially set as unity for all species. The maximum flame temperatures from these extra calculations are also shown in Fig. 1. It is found that being qualitatively consistent with the variation trend of adiabatic temperature, the peak flame temperature slightly increases as α_{CO} is increased from zero to about 0.4. Therefore, the fuel Lewis number is a key factor causing the monotonic decrease in peak flame temperature at a low strain rate. However, with further increasing α_{CO}, similar to normal calculations, the peak flame temperature also starts to decrease. This is because the addition of CO causes variation not only in the fuel Lewis number but also in chemical reactions.

The primary oxidation reaction of CO is $OH+CO=H+CO_2$. When CO is added to CH_4, the rate of this reaction is increased, and thus more OH is needed. Meanwhile, the main chain branching reaction $H+O_2=OH+O$ is also intensified because more H is formed by the previous one. The net variations of radical OH and combustion intensity depend on the balance between the two reactions when CO is added. At $a=10\ s^{-1}$, when α_{CO} becomes bigger, chemical effect causes the flame temperature to decrease, since a large amount of OH is needed to complete the reaction $OH+CO=H+CO_2$, which results in the reduction in OH concentration and combustion intensity.

With strain rate being increased to a moderate or high value, when a small amount of CO is added, the maximum flame temperature does not change (at $a=100\ s^{-1}$) or slightly increases (at $a=300\ s^{-1}$). It is because the residence time in these flames is not long enough to complete the combustion of CH_4. With the addition of a small amount of CO to CH_4, the combustion intensity is enhanced due to the reactions $OH+CO=H+CO_2$ and $H+O_2=OH+O$. This enhancement of combustion intensity does not happen at a low strain rate, since the residence time is long enough to complete the combustion of CH_4. However, when α_{CO} is increased to a larger value, the flame temperature also starts to decrease, owing to the consumption of OH by the reaction $OH+CO=H+CO_2$. This is similar to what is observed in flames of low strain rate.

Therefore, the variation of flame temperature is caused by the combination of the effects of adiabatic temperature, fuel Lewis number, chemical reaction, and residence time.

3.2 NO Formation. Figures 2 and 3 show the variations of maximum NO mole fraction and NO emission index, defined as the ratio of total formed NO to total heat release, as α_{CO} changes.

At $a=10\ s^{-1}$, the maximum NO concentration monotonically decreases with increasing α_{CO}. However, when the strain rate is increased to a moderate or high value, the maximum NO concentration first increases and then decreases. Being different, at all three strain rates, NO emission index varies in a qualitatively similar way, i.e., first increases to a critical value and then decreases, as α_{CO} is increased. However, the increase rate of NO emission index is smaller at lower strain rates. The critical CO fraction, at which NO emission index reaches its maximum, changes at different strain rates. To explain these phenomena, we first analyze the mechanism of NO formation.

Figure 4 shows the pathway of NO formation for pure CH_4 flame at $a=10\ s^{-1}$. The thickness of each line represents the magnitude of the rate and the arrow indicates the direction of the reaction. The paths with rates less than 1.0×10^{-8} mole/(cm^2 s) have been neglected. It is observed that most NO is formed by the reactions HNO $(+H,OH)\rightarrow NO$ and $N(+OH)\rightarrow NO$. Apparently, the reactions HNO $(+H,OH)\rightarrow NO$ belong to the prompt route, since species HNO is from the paths resulting from the reaction of molecular nitrogen with radical CH. Although the reaction $N(+OH)\rightarrow NO$, which was attributed to the thermal NO formation

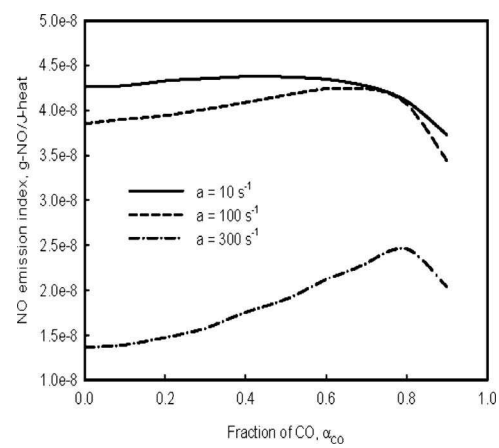

Fig. 3 Variation of NO emission index

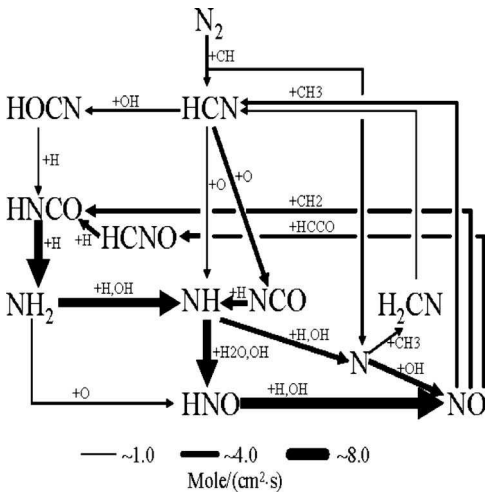

Fig. 4 NO formation pathway in CH_4/air at a strain rate of 10 s^{-1}

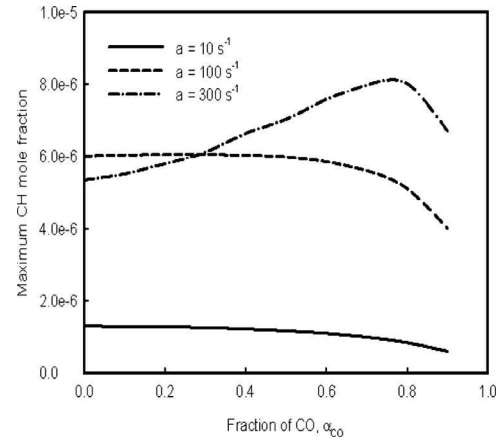

Fig. 6 Variation of peak CH concentration

route in many references, significantly contributes to NO formation, it is noted that atomic nitrogen participating in this reaction comes from the paths $N_2(+CH) \rightarrow HCN \rightarrow NCO \rightarrow NH \rightarrow N$, $N_2(+CH) \rightarrow HCN \rightarrow NH \rightarrow N$ and $N_2(+CH) \rightarrow N$. Therefore, the formation of atomic nitrogen is initiated by the reaction of molecular nitrogen with radical CH, which is the typical prompt route nitrogen conversion. Consequently, we conclude that prompt route dominates the formation of NO in a pure CH_4 diffusion flame.

In addition to the formation, NO is also consumed by reactions with some hydrocarbon radicals, such as CH_2, CH_3, and HCCO. The consumption of NO is usually called reburning. Figure 5 displays the distribution of NO formation rate for the pure CH_4 flame at $a=10\ s^{-1}$. It is found that NO is formed on the left side of stagnation plane (near the peak temperature region) and consumed later when transported to stagnation plane. Although not displayed, our results show that other flames have qualitatively similar phenomenon until the fraction of CO is increased to greater than about 0.8, i.e., prompt route dominates NO formation in most of our studied flames.

Now, we can explain the phenomena in Figs. 2 and 3. At $a=10\ s^{-1}$, the monotonic decrease in peak NO concentration is because the reduction in flame temperature (as shown in Fig. 1) reduces the formation rate of NO when CO is added. It should be pointed out that although prompt route dominates NO formation, temperature variation still modifies the rates of most reactions. However, the addition of CO moves the primary reaction zone toward the stagnation plane, which also leads to the reduction in the NO reburning rate. When a small amount of CO is added, the reduction in total NO formation (positive rate in Fig. 5) near the peak flame region is less than that of the NO reburning (negative in Fig. 5), resulting in the slight increase in NO emission index. With further increasing the fraction of CO, the significant reduction in NO formation rate results in the decrease in both peak NO concentration and NO emission index.

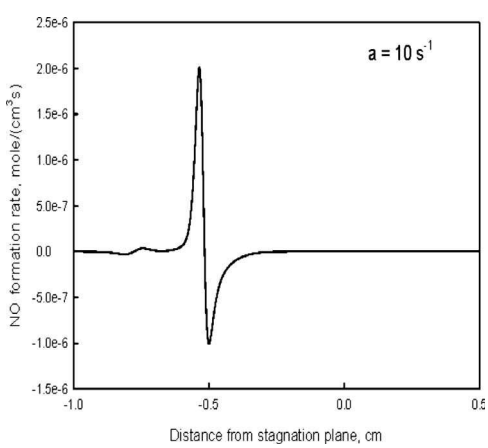

Fig. 5 Distribution of NO formation rate

When the strain rate is increased to a moderate or high value, a small amount of CO addition enhances the combustion intensity of CH_4, as mentioned before. This leads to CH radical concentration increase, as shown in Fig. 6. The increase in CH concentration and constant (at $a=100\ s^{-1}$) or increase (at $a=300\ s^{-1}$) in flame temperature cause the rise in both maximum NO concentration and NO emission index as a small amount of CO is added. This is the reason that both the maximum NO concentration and NO emission index increase when a small amount of CO is added. As noted from Figs. 2, 3, and 6, the rise rate increases with increasing strain rate. It is because the enhancement in combustion intensity is more significant at a high strain rate. With further increasing the fraction of added CO, the situation becomes similar to that at a strain rate of 10 s^{-1}, i.e., both the maximum NO concentration and NO emission index start to decrease.

Therefore, the addition of CO to CH_4 may modify the emission characteristics of NO. The effect changes at different strain rates. Overall, we find that the effect of CO addition on NO formation is minor at low to moderate strain rates. Although the addition of CO causes relatively significant increase in NO formation at high strain rate flames, the absolute NO emission level is low for these flames.

4 Conclusions

A numerical study on the effect of CO enrichment on flame temperature and NO formation in counterflow CH_4/air diffusion flames has been conducted. The results indicate that the addition of CO to CH_4 modifies both flame temperature and NO emission due to the variations in adiabatic flame temperature, fuel Lewis number, and chemical reaction. For a low strain rate flame, the addition of CO causes a monotonic decrease in flame temperature and peak NO mole fraction. However, NO emission index first slightly increases, and then decreases. When the strain rate is increased to a moderate or high value, the addition of a small amount of CO either does not change peak flame temperature too much or slightly increases it. Both peak NO concentration and NO emission index increase when a small amount of CO is added at a moderate or high strain rate. For all the cases studied, both peak flame temperature and NO emission rate start to decrease when additional CO is added.

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