The Effect of Hydrogen Enrichment on Strained Laminar Premixed and Diffusion Flames

H. Guo¹, D. Escudie²

¹. ICPET, National Research Council of Canada, 1200 Montreal Road, Ottawa, Ontario, Canada K1A 0R6
². CETHIL Centre Thermique de Lyon, UMR 5008 CNRS-INSA-UCBL, INSA de Lyon, 9 rue de la physique, 69621 Villeurbanne Cedex, France

ABSTRACT

The effects of hydrogen enrichment on flame structure, extinction and NOx formation in strained laminar premixed and diffusion flames are investigated by numerical simulation. Results indicate that hydrogen enrichment extends the lean flammability limits and thus reduces NOx formation by allowing a combustor to operate at leaner condition for premixed flames. For diffusion flames, the addition of hydrogen causes flame temperature to rise due to not only the increase in adiabatic equilibrium temperature, but also the variation in fuel Lewis number. The addition of a small amount of hydrogen has little effect on NO formation at low strain rates, and results in an increase at moderate or high strain rates. However, the addition of a large amount of hydrogen increases NO formation at all strain rates.

1. INTRODUCTION

Hydrogen enrichment is a promising concept for reducing fuel consumption and pollutant emissions. The development of hydrogen enrichment technology has motivated studies in understanding the effect of hydrogen addition on various flame properties. It has been shown that hydrogen enrichment can improve flame stability and reduce NOX formation in premixed flames [1-4], as well as increase burning velocity [5]. For diffusion combustion, hydrogen enrichment can suppress the formation of soot particles [6,7] and shorten ignition delay [8]. Relatively, not enough attention has been paid to the effect of hydrogen addition on NOX formation in diffusion flames. When hydrogen is added to a hydrocarbon diffusion flame, it is expected that the NO formation rate by the prompt route is reduced. On the other hand, the addition of hydrogen may modify flame temperature, which in turn may change NO formation rate by the thermal route. The net effect of hydrogen enrichment on NO formation in a hydrocarbon diffusion flame depends on the relative variations of the thermal and prompt routes. Naha and Aggarwal [9] investigated the effect of hydrogen addition on NOX formation in strained nonpremixed methane and n-heptane flames at a fixed strain rate (100 s⁻¹). They found that the addition of hydrogen had a minor effect on NO formation in methane flames and reduced the formation of NO in n-heptane flames. The variation in strain rate modifies the residence time of reactants in the reaction zone of a flame. The effect of hydrogen enrichment on NOX formation depends on strain rates. Therefore, it is of great interest to further investigate the effect of hydrogen addition on NOX formation in diffusion flames at various strain rates.

This paper investigates the effect of hydrogen addition to fuel on temperature and NO formation in strained laminar methane/air diffusion flames at different strain rates. For the purpose of completeness, the effect of hydrogen addition on strained laminar methane/air premixed flames is also briefly discussed.
2. NUMERICAL MODEL

The flame configuration studied is a traditional axisymmetric laminar counterflow flame. The calculations were carried out with a code as we used previously [3]. Adaptive mesh refinement is done to obtain grid independent results. Radiation heat loss is accounted for by an optically thin model [10]. The potential boundary condition is used for the free stream. The reaction mechanism used is GRI-Mech 3.0 [11]. The thermal and transport properties are obtained by using the database of GRI-Mech 3.0. The pressure and the fresh mixture temperature are 1 atm and 298 K, respectively. While hydrogen is gradually added to the fuel stream, air is maintained as the oxidant. The fraction of hydrogen, \( \alpha_{H_2} \), is defined as the volume fraction of hydrogen in the fuel stream.

3. RESULTS AND DISCUSSION

3.1. Premixed Flames

For a lean CH\(_4\)/air or H\(_2\)/air mixtures at a given equivalence ratio, there are two extinction limits when the strain rate varies [10,12]. One is caused by a high strain rate due to the shortened residence time, and the other is caused by a low strain rate because of the radiation heat loss. The former is known as the “strain extinction limit”, and the latter is known as the “radiation extinction limit”. If all the extinction limits and the corresponding equivalence ratios are plotted in a strain rate – equivalence ratio plane, a C-shaped curve is formed [3]. The upper branch of the curve is the strain extinction limit branch, and the lower branch is the radiation extinction limit branch. The region bounded by the two extinction limit branches is the flammable region of counterflow lean premixed flames. At a critical equivalence ratio, the two branches merge at one point. This critical equivalence ratio is the flammability limit of the counterflow lean premixed flame. For equivalence ratios lower than the flammability limit, a steady counterflow premixed flame cannot exist regardless of the strain rate.

Figure 1 shows the C-shaped curves of the flames with four different hydrogen fractions (\( \alpha_{H_2} \)) being 0.0, 0.1, 0.2 and 0.4, respectively. It is observed that the addition of hydrogen increases the strain extinction limit and reduces the radiation extinction limit for a given equivalence ratio. When the fraction of hydrogen is increased from 0.0 to 0.4, the flammability limit is extended from 0.420 to 0.335. As a result, with the addition of hydrogen, the flammable region of the flame is enlarged and the flammability limit is extended to a lower equivalence ratio. This allows a combustor to operate under ultra lean conditions, and thus reduce the emission of greenhouse gases, such as CO\(_2\). For example, the strain extinction limit of the
flame with the equivalence ratio of 0.5 and the hydrogen fraction of 0.4 is even higher than that of the pure CH4/air premixed flame with the equivalence ratio of 0.65, as shown in Fig.1. Figure 2 shows the NO emission indices for flames with equivalence ratio varying from 0.47 to 0.70 and the hydrogen fractions of 0.0, 0.1, 0.2 and 0.4 at a strain rate of 30 s\(^{-1}\). It is seen that at a constant equivalence ratio, the addition of hydrogen increases NO formation. However, the decrease in the equivalence ratio can significantly reduce the emission of NO. If the operating equivalence ratio of a combustor is decreased from 0.65 to 0.5 by 40% hydrogen enrichment (the example illustrated in Fig.1), the NO emission index is reduced from 0.025 to 0.007 g-NO/kg-O\(_2\). From this example, hydrogen enrichment has significantly reduced the emission of NO. Similar benefits can be demonstrated for other equivalence ratios and degrees of hydrogen enrichment.

### 3.2. Diffusion Flames

Figure 3 shows the variation of the maximum temperature at strain rates (a) of 10, 100 and 300 s\(^{-1}\), for diffusion flames. The three values were selected to represent low, moderate and high strain rates. To explain the result, the adiabatic equilibrium flame temperatures (Adia. T), obtained by equilibrium calculation at stoichiometric condition for each corresponding fuel composition, are also shown.

It is observed that the addition of hydrogen monotonically increases flame temperature at all strain rates. The first reason for this is the higher adiabatic equilibrium flame temperature of hydrogen relative to methane. However, it is noted that the rate of flame temperature increase is much faster than that of the adiabatic temperature at all strain rates, and the maximum temperatures at some \(\alpha_{H2}\) exceed the corresponding adiabatic equilibrium temperatures at a strain rate of 10 or 100 s\(^{-1}\). This phenomenon is referred to as super-adiabatic temperature. The faster temperature increase and super-adiabatic temperature occur because of the second reason, the variation in fuel Lewis number, defined as the ratio of the thermal to mass diffusion rate. It is known that for a diffusion flame, temperature is increased/decreased with decreasing/increasing Lewis number of either fuel or oxidant away from unity [13]. Ultra-low Lewis number sometimes causes a super-adiabatic temperature in a diffusion flame. The Lewis number of fuel in pure methane diffusion flames is close to unity, and hence has negligible effect on flame temperature. The Lewis number of hydrogen is significantly lower than unity. As a result, the fuel Lewis number is reduced to a value lower than unity, which tends to increase flame temperature and causes super-adiabatic temperature at some conditions as hydrogen is added. To confirm the Lewis number effect, we conducted extra calculations with the Lewis number artificially set to unity for all species at a = 100 s\(^{-1}\). These results are also shown in Fig. 3. It is found that if there were not the Lewis number effect, the
increase rate of flame temperature should have been similar to that of the adiabatic temperature, and the super-adiabatic temperature would not have occurred. Flames of other strain rates show similar results. Therefore, it is the effect of fuel Lewis number that results in the super-adiabatic temperature and the much faster increase of flame temperature than that of the adiabatic equilibrium temperature, when hydrogen is added. It is also noted that at \( a = 10 \text{ s}^{-1} \) and near \( \alpha_{\text{H}_2} = 1.0 \), the temperature increase rate slows down, and the maximum temperature becomes slightly lower than the adiabatic temperature again. This is because of radiation heat loss, which becomes more significant near \( \alpha_{\text{H}_2} = 1.0 \) due to the increased flame thickness. The impact of radiation heat loss decreases with increasing strain rate. Therefore, the decreased temperature increase does not happen at \( a = 100 \) or \( 300 \text{ s}^{-1} \), and the maximum temperatures at \( a = 100 \text{ s}^{-1} \) become higher than those at \( a = 10 \text{ s}^{-1} \) when \( \alpha_{\text{H}_2} \) is close to unity. Therefore, the effect of Lewis number significantly affects flame temperature when hydrogen is added to a methane/air diffusion flame. In the application of fuel enrichment combustion technology, this effect should be carefully taken into account.

Figure 4 displays the variations of peak NO mole fraction and NO emission index. It is revealed that at a given strain rate, although the maximum temperature monotonically increases, the behaviors of NO emission index and peak NO mole fraction are complex, as \( \alpha_{\text{H}_2} \) increases. Moreover, the variation trends differ at different strain rates. At \( a = 10 \) or \( 100 \text{ s}^{-1} \), there is a critical \( \alpha_{\text{H}_2} \) between 0.0 and 1.0, at which NO emission index reaches its maximum value. The value of the critical \( \alpha_{\text{H}_2} \) is higher at \( a = 100 \text{ s}^{-1} \) than at \( a = 10 \text{ s}^{-1} \). However, such a critical \( \alpha_{\text{H}_2} \) does not exist or becomes 1.0 at \( a = 300 \text{ s}^{-1} \). The peak NO mole fraction always reaches a maximum value at \( \alpha_{\text{H}_2} = 1.0 \), regardless of the variation in strain rate. At \( a = 10 \text{ s}^{-1} \), both NO emission index and the peak NO mole fraction are fairly constant as \( \alpha_{\text{H}_2} \) increases from 0.0 to 0.4, and then quickly increase until the maximum NO emission index is reached at the critical \( \alpha_{\text{H}_2} \). Finally, further increasing \( \alpha_{\text{H}_2} \) from the critical value to 1.0, the NO emission index decreases, while the peak NO mole fraction increases. The situation changes at \( a = 100 \) or \( 300 \text{ s}^{-1} \). When a small amount of hydrogen is added, both peak NO mole fraction and NO emission index increase. The increase is more significant at \( a = 300 \text{ s}^{-1} \) than at \( a = 100 \text{ s}^{-1} \). Then the increase of \( \alpha_{\text{H}_2} \) causes a slight decrease in both peak NO mole fraction and NO emission index. Finally, further increasing \( \alpha_{\text{H}_2} \), both parameters quickly rise until the critical \( \alpha_{\text{H}_2} \) is reached at \( a = 100 \text{ s}^{-1} \), or \( \alpha_{\text{H}_2} = 1.0 \) is reached at \( a = 300 \text{ s}^{-1} \). When \( \alpha_{\text{H}_2} \) is greater than the critical value, the variations of the two parameters at \( a = 100 \text{ s}^{-1} \) are similar to those at \( a = 10 \text{ s}^{-1} \). At \( a = 300 \text{ s}^{-1} \), they both reach their maximum at \( \alpha_{\text{H}_2} = 1.0 \).

The above phenomena are due to the variations in the mechanisms of NO formation and flame structure, when hydrogen is added. It is known that NO generally can be formed by the thermal, the prompt, the \( \text{N}_2\text{O} \) and the \( \text{NNH} \) intermediate routes in a hydrocarbon flame. The relative contribution of each route to the total NO formation can be identified by the consumption rate of molecular nitrogen by each route, as shown in Fig. 5 at \( a = 10 \) and \( 300 \text{ s}^{-1} \). A positive value means nitrogen is converted to NO or nitrogen-containing species, and a negative value indicates the opposite. For completeness, the nitrogen consumption rates by the \( \text{N}_2\text{O} \) and the \( \text{NNH} \) intermediate routes are also shown. However, their variations will not be discussed since the contributions are generally very small. It is observed that at \( \alpha_{\text{H}_2} = 0.0 \), the prompt route dominates the conversion of nitrogen at both strain rates. Alternatively, the thermal route contributes most at \( \alpha_{\text{H}_2} = 1.0 \). The combination of the variations in the nitrogen consumption rates by the thermal and prompt routes explains most of the phenomena observed in Fig. 4.

At \( a = 10 \text{ s}^{-1} \), the consumption rates of nitrogen by the thermal and prompt routes increase and decrease, respectively, leading to a nearly constant net nitrogen consumption rate and NO
emission index, as $\alpha_{H2}$ increases from 0.0 to 0.4. Further increasing $\alpha_{H2}$ from 0.4 to the critical value, the consumption rate of nitrogen by the thermal route quickly increases, while that by the prompt route gradually decreases, resulting in an increase in net nitrogen consumption rate and NO emission index. The monotonic decrease of nitrogen consumption rate by the prompt route is due to the decrease in the concentration of CH radical. For the thermal route, when $\alpha_{H2}$ increases from 0.0 to the critical value, the increase of nitrogen consumption rate is caused by the increase in temperature. When $\alpha_{H2}$ approaches unity, the final decrease in nitrogen consumption rate and NO emission index is caused by the decrease in the consumption of nitrogen by the thermal route. This will be further explained later.

At $a = 300$ s$^{-1}$, as $\alpha_{H2}$ first increases from 0.0 to a certain value, the consumption rate of nitrogen by the prompt route increases, while that by the thermal route is almost constant, leading to increases in net nitrogen consumption rate and NO emission index. At this stage, the almost constant nitrogen consumption rate by the thermal route is due to the net effects of the temperature increase that tends to raise the nitrogen consumption rate by the thermal route, and the rise in the nitrogen consumption by the prompt route that generates a large amount of atomic nitrogen and thus intensifies the forward rate of the reaction NO + N = N$_2$ + O. The increase in the consumption rate of nitrogen by the prompt route is caused by the fact that a small amount of hydrogen addition increases the concentration of CH radical, because the addition of hydrogen intensifies the combustion of a methane/air diffusion flame if the strain rate is not very low. This effect does not happen at a lower strain rate, such as at $a = 10$ s$^{-1}$, since the residence time of reactants in the reaction zone of a lower strain rate flame is long enough to complete the combustion process, and thus the addition of hydrogen only increases temperature and reduces the concentration of CH radical. With $\alpha_{H2}$ being increased beyond a certain value, the concentration of CH radical starts to decrease, resulting in a reduction in the consumption rate of nitrogen by the prompt route and NO emission index. Finally, further increasing $\alpha_{H2}$ to 1.0, the nitrogen consumption rate by the thermal route and thus the net nitrogen consumption and NO emission index increase again due to the significantly increased temperature.

The result at $a = 100$ s$^{-1}$ (not shown due to space limitation) is qualitatively similar to that at $a = 300$ s$^{-1}$, except that there is a critical $\alpha_{H2}$. When $\alpha_{H2}$ is greater than the critical value, the result at $a = 100$ s$^{-1}$ is qualitatively similar to that at $a = 10$ s$^{-1}$, i.e. the increase of $\alpha_{H2}$ reduces the nitrogen consumption rate and NO emission index, while increasing the peak NO mole fraction. This is because the reaction zone is moved further away from the stagnation plane at a higher $\alpha_{H2}$. The formation of NO in the primary reaction zone keeps increasing because of the rise in temperature, resulting in an increase in peak NO mole fraction. However, when the formed NO is transported to the region close to stagnation plane, part of the NO is converted back to molecular nitrogen by the reaction NO + N = N$_2$ + O, leading to a decrease in NO.
emission index. This phenomenon does not happen at $\alpha = 300 \text{s}^{-1}$, since the reaction zone is closer to the stagnation plane at all $\alpha_{H_2}$.

4. CONCLUSIONS

A detailed numerical study on the effect of hydrogen enrichment on strained laminar premixed and diffusion methane/air flames has been conducted. The results suggest that hydrogen enrichment extends the lean flammability limits of methane/air premixed flames. As a result, hydrogen enrichment reduces NO formation by allowing a combustor to operate at a leaner condition.

For diffusion flames, when hydrogen is added, the variations in both the adiabatic flame temperature and the fuel Lewis number significantly affect the flame temperature. Because of the Lewis number effect, the flame temperature increases much faster than the adiabatic equilibrium temperature, and a super-adiabatic flame temperature occurs at some conditions, when $\alpha_{H_2}$ is increased. Therefore, the Lewis number effect has to be carefully taken into account in the application of hydrogen enrichment technology. The addition of a small amount of hydrogen has negligible effect on NO formation at low strain rates. At moderate or high strain rates, the addition of a small amount hydrogen leads to a small increase in NO formation. Considering the fact that the absolute NO formation rate at moderate to high strain rates is usually low, we can say that the addition of a small amount of hydrogen has a minor effect on NO formation. However, the addition of a large amount of hydrogen results in an increase in NO formation.

REFERENCES