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Improvement of flame stability and NO_x reduction in hydrogen-added ultra lean premixed combustion †

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Abstract

Lean premixed combustion is a well known method in gas turbine combustors that can reduce fuel consumption and decrease flame temperature. In lean premixed flames, flame instabilities can occur because the combustion takes place near the lean flammable limit. For the purpose of increasing flame stability, a small amount of hydrogen was added into a fuel, which has ultra low lean flammable limit. The extinction stretch rate increased and total equivalence ratio at extinction decreased with hydrogen addition; consequently, ultra lean premixed combustion was possible and flame stability could be achieved at low temperature conditions. The NO_x emission increased with hydrogen addition for the same stretch rate and equivalence ratio, but the extinction stretch rate and lean flammability limit was enlarged. Consequently, NO_x emission decreased with hydrogen addition in the near extinction conditions. Hydrogen addition could improve flame stability and reduce NO_x emission in ultra lean premixed combustion.

Keywords: Extinction stretch rate; Flame stability; Hydrogen addition; NO_x emission; Ultra lean premixed combustion

1. Introduction

Combustion efficiency and clean combustion are the two main issues in recent fossil energy utilization. The control of nitrogen oxides (NO_x) has been a major issue in designing combustion systems, since NO_x play a key role in ozone depletion and the generation of photochemical smog [1, 2]. Due to the sensitivity of the NO_x production mechanism on temperature, decreasing flame temperature is a viable method in suppressing thermal NO_x formation.

There are many methods of NO_x reduction, such as flue gas dilution combustion [3, 4], lean-rich combustion, and catalytic combustion [5, 6]. Among them, lean premixed combustion is one of the promising techniques to meet the increasingly stringent NO_x emission regulations for power generation and transportation [7, 8]. Reduction of flame temperature provided by this technology can drastically reduce NO_x generation, but additional development is required to allow widespread adoption of the technology. Specifically, a better understanding of the flame stabilization is required, which is related to the fuel/air mixing and combustion temperature.

Lean premixed combustion, which has high reliability, minimal impact on turbine performance, and low emissions, is now an accepted technology for stationary gas turbine engines. However, many challenges still exist for the widespread application of lean premixed combustion for NO_x control. As NO_x emission targets continue to drop, it necessitates a lower equivalence ratio of the premixed charge. Lower temperatures of lean premixed combustion can lead to quenching of oxidation reactions, increasing CO and UHC and decreasing combustor stability [9].

To obtain lower NO_x emissions from gas turbines, a

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better understanding of the fuel/air mixing and combustion processes in practical combustors is needed. Incomplete mixing makes the local equivalence ratio exceed the overall lean equivalence ratio, resulting in hot spots within the flame and the generation of high concentrations of NO_x.

It is needed to reduce NO_x emission by maintaining lean premixed combustion system as lean as possible almost to the lean flammable limit through ultra lean premixed condition. In that case, flame instability occurs because the combustion takes place in near lean flammable limit. For the purpose of increasing flame stability, the method of adding hydrogen to hydrocarbon fuel has been tested because hydrogen has an ultra lean flammable limit. Hydrogen addition can improve combustion stability (lean flammable limit) and reduce NO_x emission simultaneously [10, 11].

We evaluated the combustion improvement and NO_x reduction characteristics in lean premixed combustion with hydrogen addition both numerically and experimentally. These researches could supply basic data for hydrogen-added combustion with lower NO_x emission, and could be applicable for the design of a very clean combustor.

2. Numerical analysis

2.1 Extinction characteristics

The characteristics of lean premixed combustion with hydrogen addition were first evaluated by numerical analysis. It was performed for methane/air premixed flame with hydrogen addition in which the total equivalence ratio (ϕ_{tot}) is 0.6. The total equivalence ratio is defined as Eq. (1). X indicates the mole fraction of each subscript species.

$$\phi_{tot} = \frac{[(X_{H2} + X_{CH4})/X_{O2}]_{actual}}{[(X_{H2} + X_{CH4})/X_{O2}]_{stoich}} \tag{1}$$

The OPPDIF counterflow code [12] based on CHEMKIN III [13] and detailed chemistry of GRI 3.0 [14] was used. Detailed governing equations are described in the OPPDIF manual [12]. Radiation and thermal diffusion effects were neglected.

The stretch rate (κ) of counterflow configuration is defined as Eq. (2).

$$\kappa = 2\frac{-u_1}{L}\left[1 + \frac{u_2}{-u_1}\sqrt{\frac{\rho_2}{\rho_1}}\right] \tag{2}$$

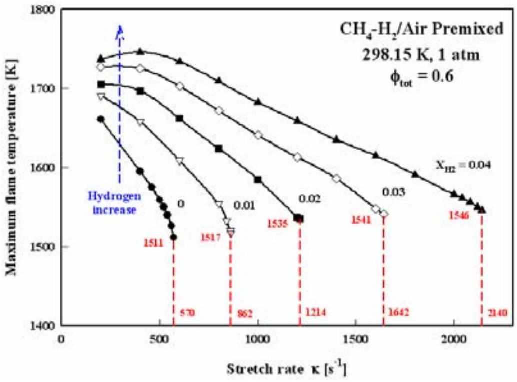

Fig. 1. Extinction S-curve for methane/air premixed flames for hydrogen addition in same total equivalence ratio (ϕ_{tot}= 0.6) condition.

u is the velocity of each nozzle and ρ is density. Subscripts *1* and *2* indicate each side of the nozzle, and L means nozzle distance. In this research, both sides of nozzles have the same composition of fuel and air mixture and exit velocity. The distance of each nozzle (L) is 2 cm.

The extinction characteristics corresponding to the upper branches of typical S-curves are shown in Fig. 1, where the maximum temperatures with stretch rate for methane/air flame with hydrogen addition are shown. Hydrogen mole fraction was varied from 0% corresponding to pure methane/air mixture to 4% in hydrogen-methane/air mixture. The calculation was conducted at room temperature and ambient pressure (T_0=298.15K, P=1atm). It showed that the maximum flame temperature gradually decreased with stretch rate and eventually the flame was extinguished. The extinction stretch rates are shown in the dashed lines.

The maximum flame temperature increased with hydrogen addition at the same stretch rate as shown in dashed arrow, which may produce a large quantity of NO emission. The extinction stretch rate was significantly increased with hydrogen addition, which means that the flame could be sustained at higher stretch rate (high scalar dissipation rate at strong turbulent intensity condition) by the hydrogen addition. The increase of extinction stretch rate can be largely attributed to the improvement of the flame speed for the CH_4/H_2 mixture in comparison to pure CH_4 [15]. The burning velocity calculated by PREMIX code [16] increased from 11.42 cm/s to 19.63 cm/s for the hydrogen mole fraction from 0% to 4%. (ϕ_{tot}= 0.6, T_0=298.15K, P=1atm).

The near extinction temperatures were slowly in

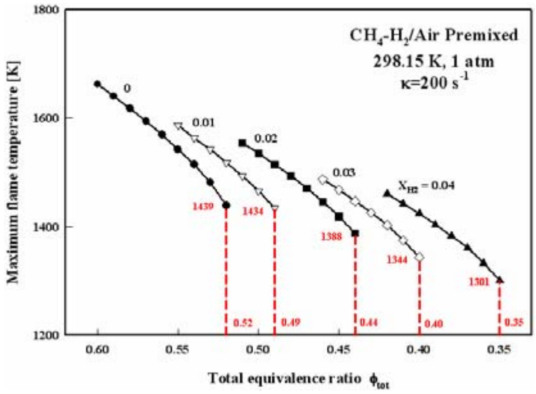

Fig. 2. Maximum flame temperature with total equivalence ration for hydrogen addition in same stretch rate (κ =200s^{-1}) condition.

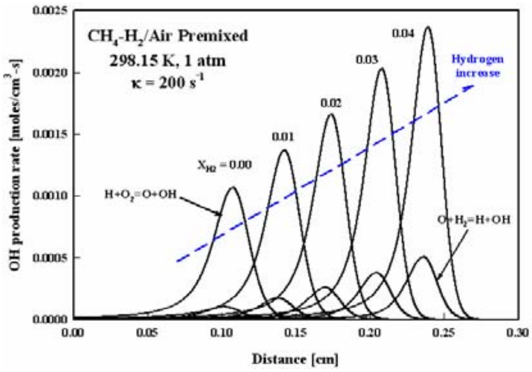

Fig. 3. OH radical production rate for hydrogen addition in same stretch rate (κ =200s^{-1}) condition.

creased but almost identical with hydrogen addition. In this calculation, the flame temperatures in all cases were below 1800K, which can be considered as the lower limit of the thermal NO_x mechanism. Therefore, it could be predicted that low NO_x production can be achieved even with hydrogen addition.

For a specified stretch rate of κ =200s^{-1}, the maximum flame temperature with total equivalence ratio is shown in Fig. 2 together with the total equivalence ratio at extinction ($\phi_{tot,ex}$) with hydrogen addition. The result indicates that the flammable limit in terms of equivalence ratio was extended from 0.52 to 0.35 with the hydrogen mole fraction of 4%, implying the possibility of ultra lean condition. The extinction temperature decreased with hydrogen mole fraction, because the extinction flame temperature of hydrogen is lower than that of hydrocarbon fuels [17]. Consequently, the flame could be sustained at lower temperature condition.

Adding the hydrogen fuel provides faster rate of H and OH radical production. Fig. 3 shows the OH production rate of two major reaction steps of $H+O_2 \leftrightarrow O+OH$ and $O+H_2 \leftrightarrow H+OH$. The result showed that the OH production rate gradually increases with hydrogen addition. The higher concentrations of the radicals may lead to the faster reaction rate and thinner reaction zone, and thereby the higher burning speed [18], which can be the reason for the improvement of flame stability by hydrogen addition.

The extension of the extinction stretch rate and lean flammable limit with the addition of hydrogen provides a strong basis for H_2 addition as a means for improving the stability of premixed flame at the temperatures below the so-called thermal NO_x threshold.

2.2 NO emission characteristics

The characteristics of NO production can be represented by the emission index of NO (EINO) following the procedure of Takeno and Nishioka [19], which is defined in terms of the NO production rate, $\dot{\omega}_{NO}$, and the fuel consumption rate, $-\dot{\omega}_F$ as shown in Eq. (3).

$$EINO = \frac{\int_{-L}^{L} \dot{\omega}_{NO} W_{NO} dx}{-\int_{-L}^{L} \dot{\omega}_F W_F dx} \tag{3}$$

where W_{NO} and W_F are the molecular weights of nitric oxide and fuel, respectively.

Fig. 4 shows the EINO with stretch rate for hydrogen mole fraction at the same value of total equivalence ratio (ϕ_{tot}=0.6). EINO gradually decreases with stretch rate in all cases due to the decrease of flame temperature. At the same stretch rate, EINO increases with hydrogen addition because the flame temperature increases with hydrogen addition as shown in Fig. 1.

Also, we evaluate the NO production mechanism for hydrogen addition, especially in near extinction stretch rate condition. The thermal and prompt NO mechanisms [20] contributing to the total EINO formation with hydrogen mole fraction are shown in Fig. 5 at the same total equivalence ratio (ϕ_{tot}=0.6) and near extinction stretch rate conditions. The thermal EINO increased with hydrogen addition because the flame temperature increased with hydrogen addition at the same stretch rate as shown in Fig. 1. And H and OH radicals increase with hydrogen addition, which may increase prompt EINO. At the same total equivalence ratio, EINO increases with hydrogen addition even in the near extinction conditions.

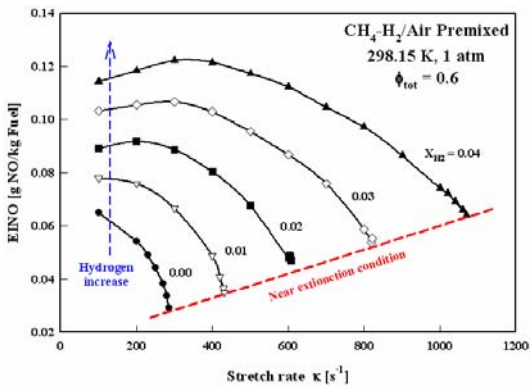

Fig. 4. EINO with stretch rate for hydrogen addition in same total equivalence ratio (ϕ_{tot}= 0.6) condition.

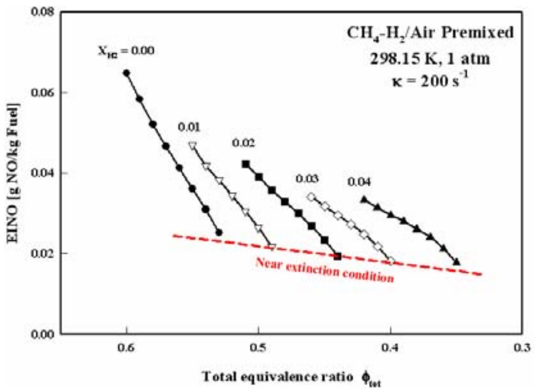

Fig. 6. EINO with total equivalence ratio for hydrogen addition in same stretch rate (κ =200s^{-1}) condition.

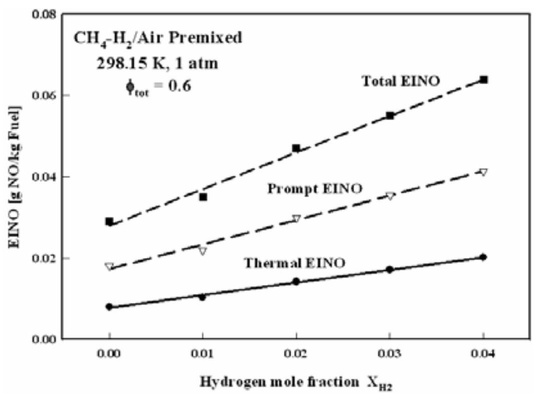

Fig. 5. Thermal and prompt EINO with hydrogen addition in same total equivalence ratio (ϕ_{tot}= 0.6) and near extinction stretch rate condition.

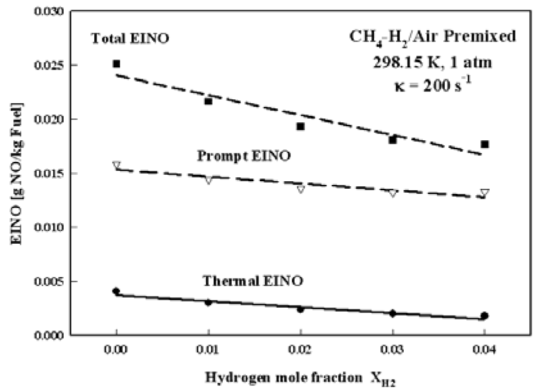

Fig. 7. Thermal and prompt EINO with hydrogen addition in same stretch rate (κ =200s^{-1}) and near extinction total equivalence ratio condition.

The EINO characteristics with total equivalence ratio at the same stretch rate (κ =200s^{-1}) are shown in Fig. 6. The EINO decreased with the decrease of total equivalence ratio, because of the lower flame temperature as shown in Fig. 2. The contribution of thermal and prompt NO mechanisms to the total EINO formation in the near extinction condition are shown in Fig. 7. The total EINO slightly decreased with hydrogen addition, which shows that the NO emission is decreased even with hydrogen addition in near extinction conditions. So the hydrogen addition could be a viable method to increase flame stability and reduce NO_x emission.

So far, the combustion characteristics with hydrogen addition have been considered by numerical analysis. In the following, the extinction characteristics and NO_x emission with hydrogen addition are evaluated by experiment for turbulent premixed flames.

3. Experimental approach

3.1 Experimental setup

The experimental apparatus consisted of a swirl burner, flow controller, a flame temperature indicator, and NO measurement setup, as schematically shown in Fig. 8.

The main fuel of methane and air for the oxidizer was supplied to each layer of the burner, and hydrogen fuel was supplied to the fuel layer. All the gases were supplied at room temperature, and flow rates were controlled by mass flow controllers (MFC, Brooks). A square duct of 250 mm x 250 mm x 600 mm with 25 mm thick insulation ceramic board was used as a furnace. Quartz glass window (200 mm x 400 mm, thickness 5 mm) was installed to visualize the flame shapes. The flame temperature was measured by R-type thermocouples along the center line of

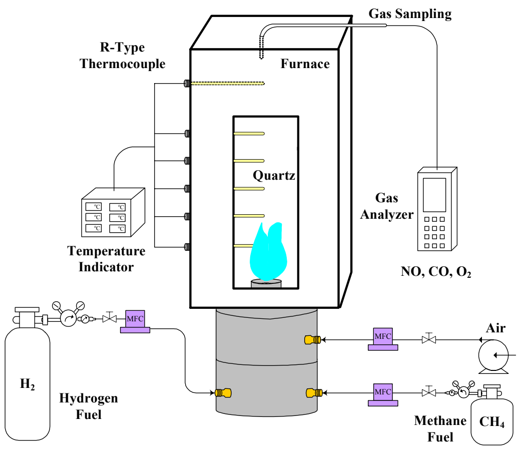

Fig. 8. Schematic diagram of experimental setup.

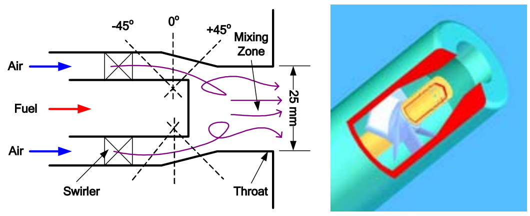

Fig. 9. Diagram of partially premixed combustor.

the furnace, and the composition of exhaust gases, such as NO, CO, and O_2 were measured by a gas analyzer (TESTO 325) at the furnace outlet.

Fig. 9 shows a schematic diagram of the partially premixed combustor. A perfectly premixed combustion with homogeneous mixture can be realized; however it faces a safety problem of flash back in practical combustion systems. Therefore, a partially premixed burner was frequently adopted for the sake of safety.

The fuel nozzle at the center of the burner had twelve holes of 1.0 mm diameter, and air was supplied through a swirler, which enhanced the mixing of fuel and air. Fuel and air were supplied from each layer and mixed in a mixing zone. To investigate the mixing characteristics, three types of fuel nozzles having different injection angles were tested -45^o, 0^o, and +45^o. After preliminary tests, the -45^o nozzle was chosen because it showed the best flame stabilization characteristics. After partial premixing, the fuel/air mixture was ejected through a 25 mm diameter nozzle into the furnace. Note that the exit velocity which is dependent on the diameter of exit nozzle could influence the flame stabilization characteristics.

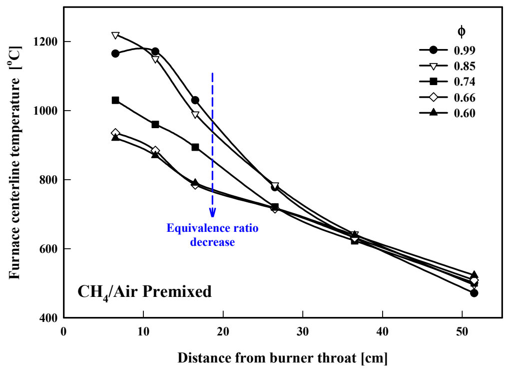

Fig. 10. Temperature profiles along centerline from throat with equivalence ratio.

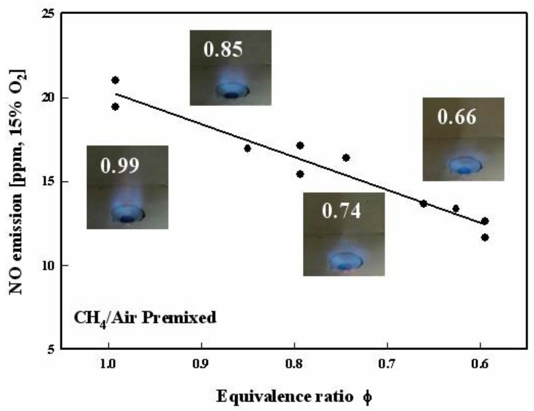

Fig. 11. Flame shape and NO emission with equivalence ratio.

3.2 Methane/air premixed combustion test

First, the characteristics of methane/air premixed flame were evaluated with the variation of equivalence ratio (ϕ), which was varied from ϕ = 0.99 to 0.6.

Fig. 10 shows the measured temperature along the centerline from the fuel nozzle for various equivalence ratios.

The temperature gradually decreases from the throat to downstream, and the temperature decreases with equivalence ratio. For ϕ=0.99, the flame was lifted and the position of the maximum temperature was away from the nozzle tip. Other cases showed that the maximum temperature appears close to the throat. Flame shape and NO emission with equivalence ratio are shown in Fig 11. The flame length decreases with the decrease of equivalence ratio, because the oxidizer flowrate was increased and a strong recirculation zone was produced by the swirler installed in the oxidizer pathway, and it made the

Table 1. Experimental conditions of hydrogen addition (variable heat capacity).

No.	Fuel (lpm)		kcal/h	X_{H2} (ϕ_{tot} = 0.6)
	CH_4	H_2		
1	12.5	0	6,420	0.000
2		1	6,575	0.005
3		2	6,730	0.009
4		3	6,884	0.013
5		4	7,039	0.017
6		5	7,194	0.020

(a) H_2 = 0 lpm (b) 1 lpm (c) 3 lpm (d) 5 lpm

Fig. 12. Photos of flame with hydrogen addition.

flame closer to the throat.

NO emission was converted into the reference value of 15% O_2 condition. The NO emission decreased with equivalence ratio, implying that the equivalence ratio should be kept as low as possible to achieve a low NO emission through ultra lean premixed combustion.

3.3 Hydrogen-added methane/air premixed combustion test

To evaluate the influence of hydrogen addition in lean premixed combustion, the methane/air premixed flame was tested with hydrogen addition. The flow rate of the main fuel methane was maintained constant (12.5 lpm) and hydrogen fuel was added varying from 1 to 5 lpm, which increased the total equivalence ratio (ϕ_{tot}). The total equivalence ratio was determined by the amount of fuel (methane + hydrogen) and air. The equivalence ratio of pure methane/air mixture was ϕ_{tot}=0.79, and it increased to ϕ_{tot}=0.87 with hydrogen addition to 5 lpm. The experimental conditions are shown in Table 1.

The flame length increased with hydrogen addition due to the increasing heat capacity and equivalence ratio. The flame color became somewhat reddish with hydrogen addition as shown in Fig. 12. The centerline temperature profile is shown in Fig. 13. The overall temperature continuously rose with the hydrogen addition because the heat capacity and total equiva-

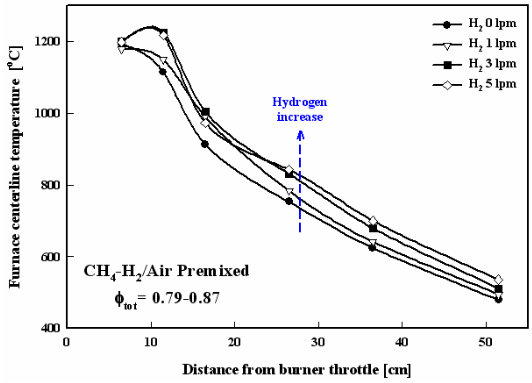

Fig. 13. Temperature profile along centerline from throat with hydrogen addition for varying equivalence ratio.

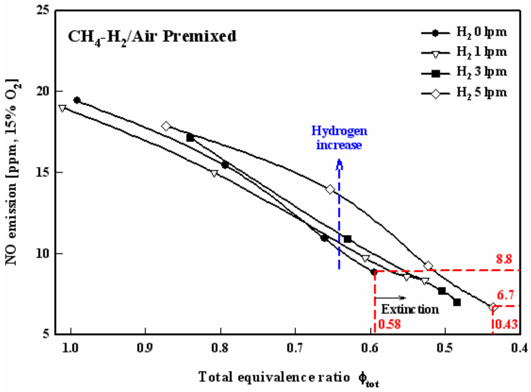

Fig. 14. NO emission with total equivalence ratio for hydrogen addition (variable heat capacity).

lence ratio were increased. It implies that hydrogen addition could produce higher NO emission.

NO emission and extinction characteristics were also evaluated with hydrogen addition. Fig. 14 shows the NO emission with total equivalence ratio for various hydrogen additions. The NO emission decreased with total equivalence ratio for all the cases of pure methane and hydrogen additions. In the case of a specified total equivalence ratio, the NO emission increased with hydrogen addition (dashed arrow) because the flame temperature was increased as previously shown in Fig. 13. Note that the equivalence ratio at extinction was extended from ϕ_{tot}=0.58 to 0.43 with hydrogen addition of 5 lpm (dashed line shows the extinction point value of NO and total equivalence ratio). This result shows that the system with hydrogen addition could be operated in ultra lean premixed condition because the flammable limit of hydrogen is very wide.

Although the NO emission increased with hydro-

Table 2. Experimental conditions of hydrogen addition (same heat capacity).

No.	Fuel (lpm)		kcal/h	X_{H2} (ϕ_{tot} = 0.6)
	CH_4	H_2		
1	12.5	0	6,420	0.000
2	12.2	1	6,421	0.005
3	11.9	2	6,421	0.009
4	11.6	3	6,422	0.014
5	11.3	4	6,423	0.018
6	11.0	5	6,424	0.022

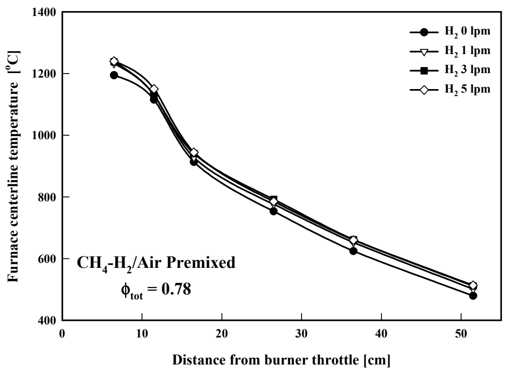

Fig. 15. Temperature profile along centerline from throat with hydrogen addition in same total equivalence ratio.

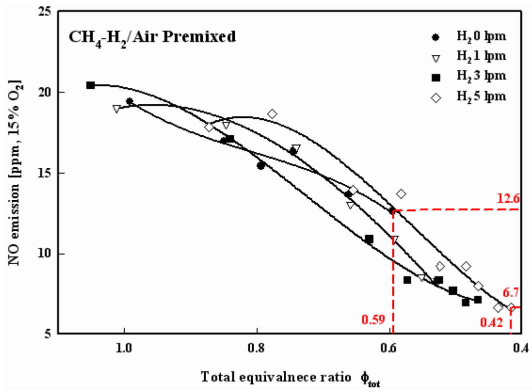

Fig. 16. NO emission with total equivalence ratio for hydrogen addition (same heat capacity).

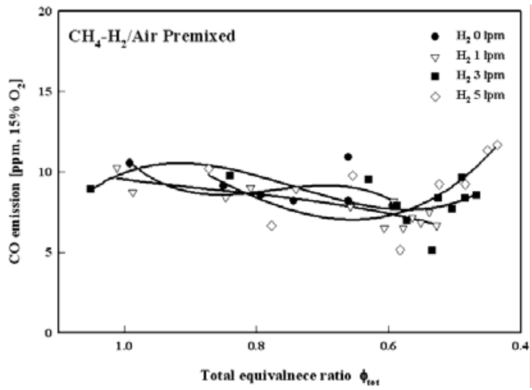

Fig. 17. CO emission with total equivalence ratio for hydrogen addition (same heat capacity).

gen addition at the same equivalence ratio, the extinction equivalence ratio decreased. Eventually, the NO emission slightly decreased with hydrogen addition form 8.8 ppm to 6.7 ppm in the range of lower flammable limit at near extinction condition. So the hydrogen addition could be a viable method to increase flame stability and reduce NO_x emission through very lean condition of operation.

However, it is difficult to compare between pure methane and hydrogen addition cases because the heat capacity can be changed with hydrogen addition. To avoid this, the case of the same heat capacity was tested. The total heat capacity was fixed at 6,420 kcal/h, which makes the flow rate of methane fuel decrease with hydrogen addition. The reference heat capacities of methane and hydrogen were 8,560 kcal/Nm^3 and 2,580 kcal/Nm^3, respectively. The experimental conditions are shown in Table 2.

The cases of the same heat capacity showed similar flame shape regardless of hydrogen mole fraction, although not shown here. Alternatively, Fig. 15 shows the centerline temperature profiles with hydrogen addition for ϕ_{tot}=0.78 case. The temperature profiles are almost the same regardless of hydrogen addition for the same heat capacity conditions, although the pure methane case shows somewhat lower than the hydrogen addition cases.

The NO emission characteristics with total equivalence ratio for the same heat capacity condition are shown in Fig. 16. The NO emission decreased with total equivalence ratio, and the effects of hydrogen addition are not shown clearly in the condition of the same total equivalence ratio. However, the total equivalence ratio at extinction was extended with hydrogen addition from ϕ_{tot}=0.59 (H_2 0 lpm) to 0.42 (H_2 5 lpm) and the NO emission was reduced by almost 50 % compared to the pure methane case at near extinction conditions. This is the same result with Fig. 14 of various heat capacity cases.

Usually, CO emission has inverse trends with NO emission, but Fig. 17 shows that the CO emission with hydrogen addition is almost the same value of about 10 ppm regardless of total equivalence ratio and hydrogen addition. Hydrogen addition may enhance the flame stability, which could restrict CO production in ultra lean premixed condition.

4. Conclusions

The following conclusions can be drawn in the ultra lean premixed combustion with hydrogen addition through the numerical and experimental approaches.

1. In the numerical calculation, the extinction stretch rate increased with hydrogen addition because the improved extinction stretch rate can largely be attributed to the increase of the flammability limit with the hydrogen addition.
2. In the numerical calculation, the total equivalence ratio at extinction decreased with hydrogen addition. Therefore, the system could be operated at ultra lean premixed condition and the flame could be stably sustained at low temperature. Also, hydrogen addition could increase OH radicals in flame, which can extend the flammable region to higher stretch rate and lower temperature condition.
3. The NO emission, which strongly depends on temperature, increased with hydrogen addition at the same stretch rate and equivalence ratio. But the extinction stretch rate and the total equivalence ratio at extinction were extended; eventually, NO emission decreased with hydrogen addition compared to near extinction conditions.
4. In the experimental test using a turbulent partially premixed burner, hydrogen addition may increase flame temperature of methane flame, but it could extend the lean flammable limit such that a reduction in NO emission can be achieved compared to the near extinction condition. In the same heat capacity condition, the flame temperature was almost identical in the conditions of the same regardless of hydrogen addition, which achieved over 50% reduction of NO emission at near extinction condition. Hydrogen addition is a viable method to enhance the flame stability and reduce the NO emission simultaneously. In the experimental results, the CO emission is almost the same regardless of hydrogen addition.

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