Effects of Addition of Carbon Dioxide on Thermal Characteristics in Oxygen-Enriched Hydrogen Flame*

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An experimental study has been conducted to evaluate the effects of CO₂ addition on thermal behavior in oxygen-enriched hydrogen flame. Experiments were performed on flames stabilized by a co-flow swirl burner, which was mounted on top of the furnace. Several different oxidizer compositions were prepared by volumetrically replacing N₂ by CO₂. In a steady state, the total as well as radiative heat flux from the flame to the wall of furnace has been measured using a heat flux meter. Temperature distribution in furnace has been also measured and compared. By increasing CO₂ proportion in the oxidizer, the convection became to play a more significant role rather than radiation due to heat blockage effect. Overall temperature in the furnace was seen to decrease, while the total heat flux has increased.

Key Words: Hydrogen Combustion, Oxygen-Enriched, CO₂, Heat Transfer, Radiation, Heat Flux Meter, NO_x

1. Introduction

Nowadays, research on hydrogen gas has been very active since it is regarded as an alternative and clean energy. Many researchers have investigated its use as a fuel in order to replace the fossil fuel, for its complete burning in air only produces H₂O and N₂. Its basic combustion characteristics have been examined by Melvin⁽¹⁾. The effect of swirl number and gas velocity on hydrogen flame height has been studied by Sano et al.⁽²⁾ On the other hand, Suzuki et al. considered the effects of additives on NO_x emission from hydrogen flame⁽³⁾.

Sometimes, oxygen-enriched combustion was introduced to raise combustion efficiency by increasing oxygen ratio in oxidizer. Oxygen-enriched combustion induces a higher flame temperature and increases the heat transfer efficiency by reducing a diluent gas of N₂ in air. It is now commercially used in the area of glass and steel production in which an environment of high temperature and radiative heat transfer is required.

Theoretically, the waste energy, that is used to heat up nitrogen in air, would be appropriated to useful energy by replacing air with oxygen. As the oxygen concentration increases, the flammability limit is also further extended

to fuel-lean side. When pure oxygen is substituted into air, the concentrations of CO₂ and H₂O become higher, which are very active in radiation so that the flame emissivity would be enhanced⁽⁴⁾. However, since the flame temperature is too high, sometimes its temperature needs to be controlled by retrieving the carbon dioxide in product gas and mixing it with reactants in regenerative way. Its effect on flame radiation in oxygen-enhanced combustion is discussed by Baukal and Gebhart⁽⁵⁾. On the other hand, some researchers have studied the effects of CO₂ as an additive to suppress NO_x for various fuel-air combustions^{(6),(7)}.

In this paper, the effect of addition of CO₂ on radiative as well as convective heat transfer has been experimentally elucidated in oxygen-enriched hydrogen flame while keeping the equivalence ratio fixed by systematically controlling a substitution of CO₂ into N₂ in air.

2. Experimental Setup

2.1 Experimental apparatus

A laboratory size of experimental furnace with burner was designed and fabricated. The furnace was vertically-downward installed to avoid full three dimensionality of the flame shape. Various gases such as hydrogen, air, oxygen and carbon dioxide, used in experiment were provided through volumetric flow meters. The signals from thermocouple and total as well as radiative heat flux meter were recorded on computer with an A/D converter board. The overall experimental setup is shown in Fig. 1.

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Fig. 1 Experimental set up

Fig. 2 Detailed drawing of furnace

2.1.1 Furnace As shown in Fig. 2, a cylindrical type of furnace has been used for experiment. Its diameter is 0.5 m and height is 1.0 m. On the surface of furnace nine ports of two different sizes were drilled to accommodate heat flux meters that are used for measuring total as well as radiative heat flux at the wall. Thermocouples were also inserted through these ports to assess gas temperature in the furnace as shown in Fig. 2. Nine holes of 2 mm diameter were also made to measure the inner wall surface temperature distribution on furnace. The Cerakwool was installed for thermal insulation inside the furnace.

2.1.2 Burner A co-annular type burner was constructed to generate hydrogen diffusion flame as seen in Fig. 3. At the tip of outer annulus of the burner is installed a swirler to stabilize flame. Outer pipe diameter for supplying oxidizer and carbon dioxide is 30 mm. Inner diameter for hydrogen is 18 mm. When carbon dioxide is added to oxidizer, a premixing-chamber is used to ensure homogeneous mixing and then supplied through outer pipe. A port for igniter is located on the inlet side of burner so that it is possible to light a fire without opening and closing upper portion of the furnace.

2.1.3 Swirler A swirler is located at the tip of outer annulus to enhance mixing rate of the oxidizer. The

Fig. 3 Detailed drawing of burner

swirler used here is of guide vane type with vane angle of 30 degrees. Following formula has been used to compute the swirl number⁽⁸⁾.

$$S = \frac{2}{3} \left[\frac{1 - (d_h/d)^3}{1 - (d_h/d)^2} \right] \tan \theta$$

where, d : inner diameter of the swirler.

d_h : hub diameter of the swirler.

θ : vane angle of the swirler.

2.2 Measurement accessories

Heat flux meters were used to check the heat transfer to the wall. These are commercial products and calibration graphs were supplied from the manufacture company.

2.2.1 Thermocouple In order to measure the temperature, two types of thermocouple were used. While K-type thermocouple was used for surface temperature of the furnace, exposed R-type thermocouple was used to measure gas temperature distribution in the furnace. Various bead sizes were used to predict real temperature by extrapolating the data to zero bead size⁽⁹⁾. Thermocouples were electrically treated for protecting them from oxidation in the flame zone. Temperature in the furnace was scanned at intervals of 1–5 cm along the radial direction and 10 cm along the axial direction.

2.2.2 Heat flux meter Commercially available heat flux sensors were used to measure the radiative and total heat transfer to the wall from the gas. It is well known Gardon type of heat flux meter.

2.2.3 Gas analyzing system Environmentally pollutant gases such as NO, CO₂, CO and SO₂ were measured by gas analyzing system. Because NO_x sensor was sensitively affected by heat, a water-cooled capturing pipe was used for extracting sample gases. Water vapor was removed by filtering before analysis.

2.3 Experimental condition

All the experimental data were retrieved when the thermal condition inside furnace reached steady state.

In this study seven experimental conditions were

Table 1 Experimental conditions

Oxygen enrichment ratio (Ω) = 0.4			
CASE	O ₂ (%)	N ₂ (%)	CO ₂ (%)
CASE 1	40	60	0
CASE 2	40	40	20
CASE 3	40	30	30
CASE 4	40	20	40
CASE 5	40	0	60
Oxygen enrichment ratio (Ω) = 0.5			
CASE	O ₂ (%)	N ₂ (%)	CO ₂ (%)
CASE 6	50	50	0
CASE 7	50	0	50

tested. For the fixed oxygen enrichment ratio of 0.4, there are five different compositions of nitrogen and carbon dioxide. Furthermore, the other two compositions were tested when the oxygen enrichment ratio is 0.5. The oxygen enrichment ratio (Ω) is defined as volumetric ratio between O₂ and total oxidizer as follows.

Ω = volume flow rate of O₂ in the oxidizer/total volume flow rate of oxidizer

Their detailed experimental compositions are listed in Table 1.

3. Experimental Results and Discussion

3.1 The effect of CO₂ on flame characteristic for case of $\Omega = 0.4$

Superficial flame behavior was observed through an observation quartz window. Originally, typical hydrogen-air flame is non-luminous and visual radiation is very small as seen in Fig. 4(a). But when the oxygen enrichment ratio becomes 0.4 without CO₂, flame emits more visual radiation so that it shows a brighter and more bluish flame as in Fig. 4(b). This is because the flame temperature increases when the oxygen concentration is enhanced. When 20% of carbon dioxide replaces nitrogen gas for the fixed oxygen enrichment ratio of 0.4, the flame emits further more visual radiation and its color turns into more white-blue as in Fig. 4(c). This is due to the fact that the carbon dioxide added plays an active role in emitting radiation. Thereafter, even if more carbon dioxide is added, the flame color doesn't make further difference.

3.2 The effect of CO₂ on temperature distribution for case of $\Omega = 0.4$

In order to examine the effect of CO₂ on temperature distribution along axial direction, the case 1, case 3 and case 5 have been selected. The case 1 corresponds to a pure oxygen enhanced combustion for $\Omega = 0.4$ without CO₂ addition. As shown in Fig. 5, the gas temperature reaches a maximum temperature of 1473°C at axial distance of 0.3 m from inlet. When the nitrogen gas is replaced by 30% of CO₂ (case 3), the maximum temperature decreases by 113°C. When 60% of CO₂ is substituted into all nitrogen gas (case 5), the maximum temperature is

Fig. 4 Visual flame characteristics

Fig. 5 Axial temperature distribution at the centerline

Table 2 Molar specific heat under constant pressure (unit: cal/mol · K)

Temperature (K)	CO ₂	N ₂
300	8.896	6.961
600	11.310	7.196
900	12.667	7.670
1200	13.446	8.061
1500	13.953	8.330
1800	14.269	8.512

reduced by 172°C. In other words, the more CO₂ gas is added to the oxidizer, the lower the overall gas temperature in upstream. Especially, the region up to 0.6 m from the inlet is affected in temperature distribution.

This phenomenon might result from a difference in thermal characteristic between N₂ and CO₂. Table 2 shows the molar specific heat of N₂ and CO₂⁽¹⁰⁾. The molar specific heat for CO₂ is bigger than that for N₂ and its difference between two gases even gets larger at higher temperature. So when CO₂ gas was supplied in place of N₂ gas on the same volumetric flow rate, the rate of increase in gas temperature is much reduced. Another reason for the lower gas temperature for more CO₂ addition is that the total heat loss to the wall is higher for more CO₂ addition as will be observed below.

Finally, there would be a supplementary reason such as chemical effects incurred by CO₂ addition as noted by Park et al. and Lee and Park^{(11),(12)}. CO is converted from CO₂ through the dominant reaction $\text{CO}_2 + \text{H} \rightarrow \text{CO} + \text{OH}$. The existence of CO inhibits the most important chain branching reaction, $\text{H} + \text{O}_2 = \text{O} + \text{OH}$ so that overall reaction rate of fuel decreases and the increase of initial CO₂ concentration induces the increase of CO mole frac-

Fig. 6 Temperature distribution along radial direction

tion. The increase of initial CO_2 concentration produces lower maximum flame temperatures.

Figure 6 shows temperature distribution along the radial direction for three cases. As seen in Fig. 6(a), the maximum temperature does not occur at the centerline, but 2 cm away from it at axial distance, $z = 0.2$ m from inlet. Its maximum temperature for case 1 is 1722°C , while it decreases by 319 and 387°C for case 3 and 5, respectively. At $z = 0.3$ m, the maximum temperature is located on the centerline again and it continuously decreases along the radial direction. At further downstream at $z = 0.6$ and 0.9 m, the radial temperature distribution becomes uniform and its temperature steadily decreases along the axial direction.

3.3 The effect of CO_2 on NO_x and CO in case of $\Omega = 0.4$

Table 3 lists the amount of NO_x formation along axial direction. As shown in table, as the amount of addition of CO_2 gas increases, NO_x formation is reduced, mainly because the maximum gas temperature is decreasing as explained above. Based on this observation, the NO_x formed

Table 3 NO_x formation at the centerline (unit: ppm)

Distance(m)	Case 1	Case 3	Case 4
at $z=0.4$	3021	1349	1247
at $z=0.6$	3008	1268	1144
at $z=0.8$	3068	1756	1178

Fig. 7 Various heat fluxes to the wall for each case

in this furnace is considered to be thermally-controlled NO_x .

CO emission is also measured using a gas analyzing system of which upper limit is 15 000 ppm. When CO_2 was added to the flame by 20% (Case 2), CO measurement is already saturated, which indicates that it exceeds 15 000 ppm. Therefore, its data is not further presented here.

3.4 The effect of CO₂ on heat flux in case of $\Omega = 0.4$

Figure 7 shows the total as well as convective and radiative heat flux toward the wall for each case. It is found that the radiative heat flux is reduced once CO₂ gas is added as shown in Fig. 7 (b) and (c) compared with Fig. 7 (a). This is considered to be due to the heat blockage effect by radiatively active CO₂ gas. In other words, the CO₂ gas absorbs the thermal radiation from the flame and re-emits much less. However, the convective heat flux is observed to increase as the addition of CO₂ gas increases. A substitution of CO₂ into N₂ based on volumetric proportion actually means that more mass CO₂ is added to the mixture gas by mass, since molecular weight of CO₂ is bigger than that of N₂. The convective heat flux is therefore, is enhanced when CO₂ is added. Thereby, the total heat flux, which is a summation of convective and radiative heat fluxes, increases as the portion of CO₂ increases.

For the case without CO₂ addition, the convection is comparable to the radiation in upstream while the radiation becomes dominant in downstream. Therefore, the radiation plays a dominant role on average for case 1. However, for the case with CO₂ addition, the convection is dominant over the radiation in upstream, while the former becomes comparable to the latter in downstream. On average the convection is predominant over the radiation when the CO₂ gas is added. Table 4 illustrates how much the total heat flux increases compared to case 1 when the CO₂ gas is added. The total heat flux on average is found to increase by 5.5 and 12% for case 3 and 5, respectively.

3.5 The effect of CO₂ on flame characteristics and NO_x for case of $\Omega = 0.5$

Two experimental conditions are considered for the oxygen enrichment ratio of 0.5 as shown in Table 1. One condition is without CO₂ in the oxidizer (case 6), while the other has 50% of CO₂ in the oxidizer instead of N₂.

Even if the oxygen enrichment ratio is increased from $\Omega = 0.4$ to 0.5, it doesn't make any difference in visual flame. But still the addition of CO₂ makes the flame brighter. Table 5 shows the NO_x formation at the centerline for case 1 and 6. When the oxygen enrichment ratio is 0.5, NO_x formation becomes more than twice as much

as that for 0.4. This is due to the higher temperature in furnace for $\Omega = 0.5$.

Likewise for the case of $\Omega = 0.4$, when CO₂ gas is added for $\Omega = 0.5$, less NO_x is detected, because CO₂ gas has the effect of reduction in gas temperature.

3.6 The effect of CO₂ on heat flux for case of $\Omega = 0.5$

As observed in Fig. 8, it shows a similar trend as for the case of $\Omega = 0.4$. When all nitrogen gas is substituted by CO₂ gas on the volumetric flow, the radiative heat flux to the wall is significantly reduced while the total heat flux increases due to the enhanced convective heat flux. Because of CO₂ gas added, the flame temperature is decreased while momentum influx from burner inlet is increasing.

For the case without CO₂ addition (case 6), the convection is comparable to the radiation in upstream while the radiation becomes dominant in downstream, which would be very similar as before with $\Omega = 0.4$. However, for the case with CO₂ addition (case 7), the convection is dominant over the radiation in upstream, and the former becomes more dominant even in downstream. Therefore, for the higher oxygen enrichment ratio, the addition

Table 5 NO_x formation at the centerline (unit: ppm)

Distance (m)	Case 1	Case 6
at z=0.4	3021	7955
at z=0.6	3008	*
at z=0.8	3068	*

* represents measurement out of limit on the gas analyzer

(a) Heat fluxes for Case 6

(b) Heat fluxes for Case 7

Fig. 8 Various heat fluxes to the wall for each case

Table 4 Increase in total heat flux compared to Case 1 (unit: %)

Axial distance (m)	Case 3	Case 5
0.1	7.82	12.52
0.2	8.05	13.64
0.3	6.34	12.24
0.4	7.53	17.50
0.5	4.89	16.50
0.6	3.90	8.39
0.7	1.64	6.97
0.8	1.29	10.99
0.9	8.29	9.99
Average	5.53	12.08

of CO₂ gas further enhances the role of convection along the downstream. The total heat flux on average is found to increase by 12.7% for case 7.

4. Summary and Conclusions

Experimental study was performed to examine the effect of CO₂ on thermal behavior in oxygen-enriched hydrogen flame. In general, a volumetric replacement of N₂ in oxidizer by CO₂ gas was found to reduce flame temperature, and thereby NO_x formation could be diminished. A decrease in flame temperature was attributed to the higher molar specific heat, higher heat loss to the wall and reduction in fuel consumption due to chemical effects.

For the oxygen-enhanced hydrogen combustion without addition of CO₂ gas, the radiation was the dominant heat transfer mode to the wall. But as the addition of CO₂ gas increased, the convection became the main heat transfer mode to the wall due to heat blockage effect in radiation, while the total heat flux increased. For the higher oxygen enrichment ratio, more NO_x was produced because of even higher gas temperature.

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