Experimental Study of the Laminar Flame Speeds of the CH₄/H₂/CO/CO₂/N₂ Mixture and Kinetic Simulation in Oxygen-Enriched Air Condition

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1. INTRODUCTION

Oxygen-enriched combustion is a promising technology to reduce pollution. This technology uses oxygen-enriched air or pure oxygen instead of air,¹ which improves environmental protection significantly.² Because of the increase of oxygen content, the flue gas contains a lot of high concentration carbon dioxide, which is helpful for capturing carbon. The volume of the flue gas is reduced in the condition of high O₂ concentration because of the decrease of nitrogen content, which reduced the heat loss of the flue gas. On the other hand, the proportions of CO₂ and H₂O in the combustion products increase, so the radiation capacity of the products is greatly improved compared with that of traditional combustion. Thus, combustion efficiency and radiative heat transfer are improved by increasing oxygen content. Some fuels with low calorific value can reach higher combustion temperatures.

The laminar flame speed is an important parameter for studying flame characteristics.³,⁴ The enriched oxygen content in the oxidizer changes the combustion characteristics, including the laminar flame speed. There are a lot of research studies that focus on the combustion characteristics of fuels in oxygen-enriched air condition. Mazas et al.⁵ measured the laminar flame speed of CH₄/O₂/N₂/H₂O. They found that the increasing O₂ content increased the laminar flame speed, while the chemical effect of H₂O is reduced. Similar conclusions were obtained by Chica Cano et al.⁶ Takahashi et al.⁷,⁸ studied the extinguishing features of the CH₄ diffusion flame, which is changed by the O₂ concentration. Zhou et al.⁹ studied the effects of O₂ and CO₂ on the laminar burning velocity of the O₂−N₂−CO₂−iso-octane mixture. Results show that the added CO₂ and increasing O₂ content changed the laminar flame speed significantly. Pio et al.¹⁰ studied the laminar flame speeds of CH₁/O₂/air experimentally. They found that the increase of O₂ content promoted the laminar propagation velocity and expanded the flammable range of CH₁. Salzano et al.¹¹ and Xu et al.¹² studied the explosive characteristics of syngas and hydrous bioethanol in oxygen-enriched air conditions. Results show that the rising oxygen fraction promoted the deflagration index and explosive pressure. Boushaki et al. investigated the effect of the added O₂ on the generation of NO, NOₓ, and CO₂.¹³ The results show that oxygen enrichment is favorable for the increase of CO₂ and NOₓ and the decrease of CO emissions. Alabas B et al.¹⁴ found that the oxygen enrichment of air reduces the CO and increases the NO generation for the H₂/CO/CH₄ mixture in a swirl burner.

Generally, the laminar burning velocity is affected by the compositions of fuels,¹⁵,¹⁶ oxygen concentration,¹⁷,¹⁸ dilution gas,¹⁹ initial temperature,²⁰ and pressure.²¹ For example, Nair et al.²² studied the laminar flame propagating velocity of liquid petroleum gas in air condition. Weng et al.²³ measured...
the laminar flame speed of $\text{H}_2$ in $\text{O}_2/\text{N}_2$ and $\text{O}_2/\text{CO}_2$ atmosphere using a Bunsen flame. Yelishala et al.\textsuperscript{27} investigated the effects of the equivalence ratios, the initial pressure, and temperature on the laminar flame propagating velocities of propane/air/CO\textsubscript{2} mixture. Wu et al.\textsuperscript{28} measured the laminar flame propagation velocity of the CH\textsubscript{4}/air and the acetone/air mixture in a closed vessel. All of the above research shows that the laminar flame speeds of a gas mixture are affected significantly by the compositions of the gas mixture. The laminar flame speed of fuel in oxygen-enriched air condition is different from the air atmosphere because the high oxygen content of the mixture changes the combustion process.

In fact, there are a large number of fuels with a low calorific value in the industrial field. A large number of gas fuels, such as the blast furnace gas, are produced in the process of iron-making in iron-steel plants. The compositions of the blended gas produced in steel and iron plants usually consist of CO, H\textsubscript{2}, and CH\textsubscript{4}, while the inert gas CO\textsubscript{2} and N\textsubscript{2} were mixed in the blended gas. If the oxygen-enriched combustion technology can be applied to the blended gas in iron-steel plants, it will reduce a large amount of energy consumption and CO\textsubscript{2} emission. However, the studies on the laminar flame speeds of the blended gas in oxygen-enriched air are inadequate, especially for the complex blended gas. The laminar flame speeds of the blended gas are very useful for industrial applications, such as the blended gas burner design and safety prediction. In the present work, the laminar flame speeds of the blended gas were studied in oxygen-enriched air, which is helpful for the application of the blended fuel in the iron-steel field in oxygen-enriched air condition. The laminar flame speeds of the blended gas are new and the discussion about the factors on the laminar flame speed is useful for designing new burners or any other purpose.

The main components of the blended gas are CO, H\textsubscript{2}, CH\textsubscript{4}, CO\textsubscript{2}, and N\textsubscript{2}. The concentration of each species is shown in Table 1. Besides the species in Table 1, there is a very low proportion of O\textsubscript{2} (less than 1.5%). Here, the residual O\textsubscript{2} was considered in the oxidizer ($\text{O}_2/\text{N}_2$).

| species     | CO  | H\textsubscript{2} | CH\textsubscript{4} | CO\textsubscript{2} | N\textsubscript{2} |
|-------------|-----|-------------------|-------------------|-------------------|-------------------|
| volume concentration (%) | 17.34 | 24.55 | 9.30 | 10.87 | 36.56 |

2. EXPERIMENTAL METHODS

Figure 1 shows an experimental device, which consists of a gas supply system, a constant temperature water bath, a Bunsen burner, and a digital camera (Canon, 80D). The pure CO, $\text{H}_2$, CH\textsubscript{4}, CO\textsubscript{2}, O\textsubscript{2}, and N\textsubscript{2} came from the high-pressure gas cylinders. The purity of each species is above 99.99%. Six mass flow controllers (MFCs) were used to control the flow rate of each species. After MFC, six kinds of gases were mixed in a pipe sufficiently. The pipe is over 10 m long to mix the gas mixture thoroughly. Then, the blend gas was preheated in the water bath. The temperature of the water bath was controlled by the temperature controller. Finally, the premixed gas entered the burner, which was made of a straight tube. Three stainless steel tubes with 6, 8, and 12 mm inner diameter, respectively, were used to meet the requirement of different flow rates. The length of each stainless steel tube is about 1.0 m, which ensures that the laminar flow of the gas mixture is fully developed. A digital camera was used for recording the flame pictures. The flame contour was captured and the area of the flame surface (reaction zone) was calculated using these flame pictures. The experimental measurements were performed at 300 K, 1 bar.

Based on the mass balance, the formula for calculating the laminar flame speed $S_L$ of a one-dimensional laminar flame is as follows.\textsuperscript{29,30}

$$S_L = \frac{\dot{Q}}{A_b}$$

where $A_b$ is the area of the reaction zone and $\dot{Q}$ is the total volume flow rate of the premixed unburned gas. The value of $\dot{Q}$ was the sum of the values of six MFCs, and $A_b$ was obtained from the flame picture. The shape of the flame is a cone, and the area of the reaction zone is determined by the edge of the cone. The edge of the reaction zone is the position where the flame temperature is the highest. The brightest position in the vertical direction of the flame image corresponds to the hottest point of the flame. First, the brightest points in the flame image were captured using a piece of Matlab code, then the value of $A_b$ was calculated based on the captured edge of the reaction zone.

The uncertainty of the measurement is mainly from two aspects. On the one hand, the uncertainty is from the experimental method, including the heat loss from the reaction zone to the unburned gas, the calculation of the flame area, and the estimation of $S_L$ using eq 1. The uncertainty in this aspect is about 3% concerning all of the influence factors. On the other hand, the uncertainty is from the error of the instruments, including the errors of the MFCs and the temperature fluctuation. The total error from the MFCs is about 3% and that from the temperature is about 1.5%. Thus,
the total uncertainty of the measurement is expected to be 4.5% (eq 2).

\[
\sqrt{3^2 + 3^2 + 1.5^2} = 4.5\%
\] (2)

3. COMPUTING METHODS

In the present work, the PREMIX code\(^\text{31}\) in Chemkin package was employed to calculate the laminar flame speed. A freely propagating flame was chosen in the kinetic simulation. In the calculation, the maximum grid number was set to be 400. Both the GRAD and CURV values were 0.01. The length of the reaction zone was 10 cm to ensure that the chemical reactions have been completed in the reaction zone. For comparison, the GRI Mech 3.0\(^\text{32}\) and USC Mech II mechanism\(^\text{33}\) were used in the calculation. GRI Mech 3.0 was usually used for describing methane oxidation. This mechanism contains 53 components and 325 elementary reactions. The USC II mechanism was proposed for describing the combustion of CO/H₂/hydrocarbon fuels (C\(_{1-4}\)). This mechanism contains 111 components and 784 elementary reactions.

4. RESULTS AND DISCUSSIONS

4.1. Effect of the Equivalence Ratios. Figure 2 shows the measurements and calculations of \(S_L\) of the blended gas versus the equivalence ratios \(\phi\) at different oxygen concentrations (21, 25, 29, 33%). Here, the oxygen concentration is the mole fraction of O\(_2\) in the oxidizer (O\(_2\)/N\(_2\)). As seen in Figure 2, both the experimental and calculated \(S_L\) increased with the increase in the equivalence ratio at the lean-fuel side and decreased at the rich-fuel side. The peak value of \(S_L\) appears at the equivalence ratio from 1.0 to 1.2. The peaks of the experimental laminar flame speeds are 43.6, 62.1, 77.3, 87.2, and 102.7 cm/s at 21, 25, 29, 33, and 37% oxygen concentrations, respectively. Figure 3 shows the measured laminar flame speeds of pure fuels and syngas in air condition. Through comparison, the peak position of the blended gas is close to that of pure CH\(_4\), C\(_2\)H\(_6\), and different from that of syngas.\(^\text{26}\) The peak position of the maximum \(S_L\) of H\(_2\)/CO mixture appears at around \(\phi = 2.4\) because the content of CO (75%) in the H\(_2\)/CO mixture is much higher than the H\(_2\) content (25%). These experiments indicate that the position of the peak value is closely related to the composition of fuels. The fractions of H\(_2\) and CH\(_4\) are higher than that of CO in the blended gas, thus the change trend and the position of the peak value of the blend gas are mainly determined by the content of H\(_2\) and CH\(_4\).

The effect of the equivalence ratio on the \(S_L\) is mainly due to the change in the flame temperature. As the equivalence ratio increased from the lean-fuel to the rich-fuel sides, the flame temperature increased first and then decreased. Near the stoichiometric point (\(\phi = 1.0\)), the fuel and oxidizer are completely consumed, and the flame temperature reaches the maximum value. Thus, the peak point of the laminar flame speed appears near the stoichiometric point.

It is found that \(S_L\) has a nearly quadratic function relationship with the equivalence ratio. Through comparison, the measured laminar flame speeds are found to be close to the calculated results using GRI 3.0. The average difference between the measurements and the calculation with GRI 3.0 is about 3%, while the average difference is about 7% for USC II. Thus, the discussions were performed with the calculated results using GRI 3.0 in the following sections.

4.2. Effect of Oxygen Concentration. Figure 4 shows \(S_L\) versus oxygen concentrations in some cases. The results show that under a certain equivalence ratio, \(S_L\) changes approximately linearly with increasing oxygen concentration, which is different from the change trend of some fuels. For example, \(S_L\)
of methane has a “u” (approximately quadratic function) relationship with oxygen concentrations.

The effects of the oxygen concentration on $S_\alpha$ are from two sources. On one side, the increasing oxygen concentration reduces the $N_2$ content, which decreases the amount of the exhaust and reduces the heat loss significantly. Thus, the adiabatic flame temperature is promoted by the increasing oxygen concentration. On the other side, the $O_2$ molecule
enhances the free radical content in the reaction zone, such as O radical, which increases the collision probability of free radicals and promotes the branch reactions.

Figures 5–7 show the reaction pathway of the blended gas in different oxygen concentrations (21, 25, and 29%). In these figures, the arrow indicates the oxidation process of the major intermediates. The reaction numbers were marked along the arrows. The numbers in the brackets are the contribution rates of the related reactions. CO is an important intermediate in the oxidation process of CH₄ because the oxidation process of CO is involved in that of CH₄. H₂ is an important combustible species in the blended gas. Thus, the reaction pathway of the blended gas consists of the oxidation process of CH₄ and H₂. It is noted that several “CO” and “H₂” are used in these figures to express the pathway more clearly. Here, these “CO” or “H₂” are the same substance.

The major pathway of the oxidation of CH₄ is as follows: CH₄ → CH₃ → CH₂O → HCO → CO → CO₂, which is consistent with our previous results. This major oxidation pathway is not affected by the increasing oxygen concentration, but some contribution rates of element reactions are changed by elevated oxygen concentration. There are three major element reactions in the first step of dehydrogenation of CH₄: O + CH₄ ↔ OH + CH₃ (R11), H + CH₄ ↔ CH₃ + H₂ (R53), and OH + CH₄ ↔ CH₃ + H₂O (R98). Most of CH₄ is oxidized into CH₃ through the three reactions. The sum of the contribution rates of these elementary reactions is 98–99%, which is not changed by the oxygen concentration. H + CH₃ (+M) ↔ CH₄ (R52) is the most important reaction for CH₄ generation. This reaction is also an important elemental reaction that decreases Sₗ because of the prevention of CH₄ dehydrogenation.

The largest H₂ consumption is from OH + H₂ ↔ H + H₂O (R84). This reaction is the major pathway to generate H radical and H₂O from H₂ oxidation. Because of the generation of H, this reaction plays an important role in promoting Sₗ. However, the contribution rate of this reaction is slightly reduced by the rise of O₂ concentration, which is due to the competition of H₂ with other radicals such as O. The second important reaction for H₂ oxidation is O + H₂ ↔ H + OH (R3). This reaction increases H and OH, which increases Sₗ. The contribution rate of this reaction was enhanced with the increased oxygen concentration. Besides the addition from the blended gas, H₂ is generated from the CH₄ dehydrogenation through the reaction H + CH₄ ↔ CH₃ + H₂ (R53), H + CH₄ ↔ CH₃ + H₂ (R45), H + CH₂O ↔ HCO + H₂ (R58), and O + CH₃ ↔ H + H + CO (R284). The contribution rates of these reactions are also changed by the oxygen concentration, but both the generation and consumption pathways of H₂ are changed little by the oxygen concentration.

Almost all of the CO was oxidized into CO₂ through the reaction OH + CO ↔ H + CO₂ (R99), which has a more than 97% contribution rate. However, the effect of R99 was decreased by the increase in oxygen concentration. Because the contribution rate of the reaction O + CO (+M) ↔ CO₂ (+M) (R12) was enhanced by the increasing O, which was closely related to O₂ concentration.

4.3. Influence of the Composition of the Blended Gas. The blended gas composed of combustible components (H₂, CO, CH₄) and incombustible components (N₂ and CO₂). The effects of each species on Sₗ are discussed in this section. Here, the concentration of each species is the volume fraction of the species in the blended gas. If one component concentration increases, the concentrations of other components are reduced in the initial proportions.

Figure 7. Reaction pathway of the blended gas at 29% oxygen concentration.
4.3.1. Effect of $H_2$, CO, and $CH_4$. Figure 8 shows $S_L$ of the blended gas versus the $H_2$ concentration at the stoichiometric ratio ($\phi = 1.0$). The results show that $S_L$ was enhanced by the increase in $H_2$ concentration from 0 to 60%. The rate of increase in $S_L$ increased with an increase in $H_2$ content. The increase in $H_2$ concentration promotes the calorific value of the blended gas. Figure 8 also displays variation in the adiabatic flame temperature versus $H_2$ concentration. The equilibrium gas model coupled with the detailed thermal data of GRI 3.0 is used to estimate the adiabatic flame temperature. The result shows that the increasing $H_2$ concentration increased the adiabatic flame temperature because the combustion heat release was increased by $H_2$. The increasing $H_2$ increased the H and OH radicals through reactions such as R3, which accelerates the combustion rate.

Figure 9 shows the change in $S_L$ versus CO concentration. The laminar flame speed was increased with the rise in the combustion release from increasing CO. However, the increased rate was reduced with the increasing CO until it became negative in the condition of high CO concentration. On the one hand, the calorific value of CO is lower than that of $H_2$ and $CH_4$. The concentrations of $H_2$ and $CH_4$ decreased with the increasing CO, so the increased rate of the total heat release of the blended gas was reduced. On the other hand, the amount of some free radicals such as H decreased with the decrease of $H_2$ and $CH_4$ content, because the free radicals containing H were generated from the dehydrogenation. The decrease in free radicals slowed down the laminar flame speed.

Figure 10 shows that $S_L$ of the blended gas decreased with the increase in the $CH_4$ concentration from 0 to 60%. The decrease in $S_L$ under the influence of $CH_4$ was because of the increase in the $N_2$ concentration in the oxidizer ($O_2/N_2$). For example, under the condition of 30% oxygen concentration, the chemical reaction equations of $CH_4$, CO, and $H_2$ were as follows.

$$CH_4 + 2O_2 + 4.66N_2 = >CO_2 + 2H_2O + 4.66N_2$$ (3)

$$CO + 0.5O_2 + 1.17N_2 = >CO_2 + 1.17N_2$$ (4)

$$H_2 + 0.5O_2 + 1.17N_2 = >H_2O + 1.17N_2$$ (5)

According to the above equations, about 4.66 mol $N_2$ is added in the premixed mixture from the oxidizer for 1 mol $CH_4$, while 1.17 mol $N_2$ is added for 1 mol $H_2$ or CO. It is inferred that there is more $N_2$ in the combustion process of $CH_4$ than that of $H_2$ or CO. $N_2$ is an inert gas that reduces the laminar flame speed through the thermal and dilution effects. The adiabatic flame temperature of the blended gas is promoted by the $CH_4$ content, which indicates that the thermal property of the premixed gas is not the major reason for the decrease of $S_L$. Thus, the dilution effect of $N_2$ is the main factor in the reduction of $S_L$. Two $N$ atoms connected with each other by bonds in the $N_2$ molecule. The $N_2$ molecule acts as a barrier to isolate the free radicals, the fuel, and $O_2$. Thus, the reaction rates and the laminar flame speeds are decreased by the elevated $CH_4$ concentration.

4.3.2. Effect of $N_2$ and $CO_2$. Figures 11 and 12 show the effects of $N_2$ and $CO_2$ concentrations on $S_L$, respectively. The increase in $N_2$ and $CO_2$ concentrations decreases the laminar flame speed. This is mainly because both $N_2$ and $CO_2$ are incombustible species. First, the increase in $N_2$ or $CO_2$ content reduces the amount of $H_2$, $CO$, and $CH_4$, which decreases the total heat release of the blended gas. Second, the dilution effect of $N_2$ or $CO_2$ reduces the collision possibility of free radicals. Third, the added $N_2$ or $CO_2$ decreased the adiabatic flame temperature through the thermal property. Through comparison, the inhibition effect of $CO_2$ on SL is bigger than that of $N_2$. On the one hand, the specific heat capacity of $CO_2$ is higher, thus the flame temperature of the gas mixture diluted by $CO_2$ is lower. On the other hand, the $CO_2$ molecule has a larger diameter and a larger surface area. $CO_2$ reduces more possibility of collision between active molecules. In addition, $CO_2$ is not an inert gas but participates in chemical reactions,
such as R99, which reduces the H concentration and combustion reaction rate.\textsuperscript{36} For the above reasons, the inhibition effect of CO\textsubscript{2} on $S_L$ of the blended gas is bigger than N\textsubscript{2}.

5. CONCLUSIONS

In this paper, $S_L$ of the CH\textsubscript{4}/H\textsubscript{2}/CO/CO\textsubscript{2}/N\textsubscript{2} mixture has been investigated using a Bunsen flame and the kinetic simulation. These results are very useful for the oxygen-enriched combustion technology of the flexible blended gas with low calorific value in the industrial field. The following conclusions are formulated from the experiments and calculations. First, the peaks of the experimental laminar flame speed of the CH\textsubscript{4}/H\textsubscript{2}/CO/CO\textsubscript{2}/N\textsubscript{2} mixture are 43.6, 62.1, 77.3, 87.2, and 102.7 cm/s at 21, 25, 29, 33, and 37% oxygen concentrations, respectively, which usually appeared at the equivalence ratio from 1.0 to 1.2. Second, the elevated oxygen concentration promoted the laminar flame speed. Third, the reaction rates and the contribution rates of the reactions were changed by the increased oxygen concentration of an oxidizer, but the reaction pathway changed little. Next, the laminar flame speed increased with the increase in H\textsubscript{2} and CO concentrations but decreased with the increase in CH\textsubscript{4}, CO\textsubscript{2}, and N\textsubscript{2} contents. Lastly, the inhibition effect of CO\textsubscript{2} on $S_L$ is bigger than that of N\textsubscript{2}, which is due to the higher specific heat and chemical effect of CO\textsubscript{2}.

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\subsection*{Notes}

The authors declare no competing financial interest.

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