

EFFECTS OF HYDROGEN AND CARBON DIOXIDE ON THE LAMINAR BURNING VELOCITIES OF METHANE—AIR MIXTURES

Ueda, A.¹, Nisida, K.², Matsumura, Y.³, Ichikawa, T.⁴, Yutaka Nakashimada, Y.⁵, Endo, T.⁶, Johzaki, T.⁷ and Kim, W.⁸

1 Department of Mechanical Systems Engineering, Hiroshima University, 1 Chome-3-2 Kagamiyama, Higashihiroshima, Hiroshima Prefecture 739-8511 Japan, g210085@hiroshima-u.ac.jp

2 Department of Mechanical Systems Engineering, Hiroshima University, 1 Chome-3-2 Kagamiyama, Higashihiroshima, Hiroshima Prefecture 739-8511 Japan, nishida@hiroshima-u.ac.jp

3 Department of Mechanical Systems Engineering, Hiroshima University, 1 Chome-3-2 Kagamiyama, Higashihiroshima, Hiroshima Prefecture 739-8511 Japan, mat@hiroshima-u.ac.jp

4 Department of Mechanical Systems Engineering, Hiroshima University, 1 Chome-3-2 Kagamiyama, Higashihiroshima, Hiroshima Prefecture 739-8511 Japan, tichi@hiroshima-u.ac.jp

5 Department of Mechanical Systems Engineering, Hiroshima University, 1 Chome-3-2 Kagamiyama, Higashihiroshima, Hiroshima Prefecture 739-8511 Japan, nyutaka@hiroshima-u.ac.jp

6 Department of Mechanical Systems Engineering, Hiroshima University, 1 Chome-3-2 Kagamiyama, Higashihiroshima, Hiroshima Prefecture 739-8511 Japan, takumaendo@hiroshima-u.ac.jp

7 Department of Mechanical Systems Engineering, Hiroshima University, 1 Chome-3-2 Kagamiyama, Higashihiroshima, Hiroshima Prefecture 739-8511 Japan, tjohzaki@hiroshima-u.ac.jp

8 Department of Mechanical Systems Engineering, Hiroshima University, 1 Chome-3-2 Kagamiyama, Higashihiroshima, Hiroshima Prefecture 739-8511 Japan, kimwk@hiroshima-u.ac.jp

ABSTRACT

The effects of different mole fractions of hydrogen and carbon dioxide on the combustion characteristics of a premixed methane–air mixture are experimentally and numerically investigated. The laminar burning velocity of hydrogen-methane-carbon dioxide-air mixture was measured using the spherically expanding flame method at the initial temperature and pressure of 283 K and 0.1 MPa, respectively. Additionally, numerical analysis is conducted under steady 1D laminar flow conditions to investigate the adiabatic flame temperature and dominant elementary reactions. The measured velocities correspond with those estimated numerically. The results show that increasing the carbon dioxide mole fraction decreases the laminar burning velocity, attributed to the carbon dioxide dilution, which decreases the thermal diffusivity and flame temperature. Conversely, the velocity increases with the thermal diffusivity as the hydrogen mole fraction increases. Moreover, the hydrogen addition leads to chain-branching reactions that produce active H, O, and OH radicals via the oxidation of hydrocarbons, which is the rate-determining reaction.

NOMENCLATURE

A	Flame area	X_{H_2}	Hydrogen mole fraction
l	Spatial distance	Z_{CO_2}	Dilution ratio of carbon dioxide
L_b	Burned gas Markstein length	α	Thermal diffusivity
n_i	Mole number of each gases	δ	Laminar flame thickness
r	Flame radius	K	Flame stretch rate
S_n	Flame propagation speed	ρ_b	Burned gas density
S_n^0	Stretched laminar flame speed	ρ_u	Unburned gas density
S_u^0	Unstretched laminar burning velocity	σ	Expansion ratio
T_{ad}	Adiabatic flame temperature	ϕ	Equivalence ratio
x_i	Mole fraction of each radicals		

1. INTRODUCTION

Recently, there has been a growing interest in renewable energy as a new clean energy alternative to fossil fuels to prevent global warming. In particular, power generation using biogas has been attracting attention because it is generated from discarded food waste and livestock waste, unlike wind power or solar power generation, which use natural phenomena to generate electricity. Consequently, we expect a stable supply of energy that is not affected by the weather. In the biogas production process, microorganisms decompose the organic matter in the materials, after which methane is extracted. In contrast, during microbial fermentation, carbon dioxide and methane are produced simultaneously. The dilution of gases affects the combustion characteristics, such as the laminar burning velocity, flame temperature, thermophysical properties, and chemical reactions, and this effect on the combustion phenomena varies depending on the gas species that dilute the mixture [1-3]. For instance, if the mixture is diluted with a gas with a small specific heat at low pressure, such as argon, the flame temperature and the laminar burning velocity increase [4]. Previous studies have emphasized that the dilution of carbon dioxide leads to a decrease in the laminar burning velocity [5-14] and flame temperature [5, 6, 9, 15]. This is because the carbon dioxide is a nonflammable gas; thus, the dilution of carbon dioxide will reduce the concentration of the flammable gas in the mixtures. In addition, carbon dioxide requires more energy to raise the temperature than nitrogen because it has a larger specific heat than those of other gases. The decrease in the laminar burning velocity and flame temperature reduces the amount of energy that can be obtained by methane combustion, which decreases the energy efficiency of the biomass fuel. The addition of hydrogen to a mixture of methane and carbon dioxide is expected to be an important strategy for solving these problems because it has a faster laminar burning velocity and a higher flame temperature than those of methane [16-21]. The fuel mixture of hydrogen and methane is called Hythane, and it is expected to be an alternative clean energy source [22, 23]. The effect of hydrogen addition to methane on the combustion characteristics has been demonstrated in previous studies to

increase the laminar burning velocity [11, 14, 18, 19, 21, 24-31]. Given the above considerations, the objective is to clarify the improvement mechanism of the laminar burning velocity when a small amount of hydrogen is added to the mixture, without the any noticeable effect. The laminar burning velocity of hydrogen-methane-carbon dioxide spherically propagating flame was experimentally measured.

2. EXPERIMENTAL AND COMPUTATIONAL SPECIFICATIONS

The experimental facility or set up which consists of a high-pressure chamber with a total volume of 0.79 L, an ignition system, and a high-speed schlieren image system was used to measure the laminar burning velocity of the premixed gas explosion. A schematic of the experimental setup is shown in the literature [32]. In the experiment, the vessel was vacuumed and filled with a hydrogen–methane–carbon dioxide–air mixture according to the corresponding partial pressure. After that, the mixture was ignited by centrally located two electrodes, equipped on both sides of the container and linked to a high-voltage source. The behaviors of flame propagations were filmed using the schlieren image system with a high-speed camera at 5000 frames per second.

The equivalence ratio of the hydrogen–methane–carbon dioxide–air mixture is given by

$$\phi = \frac{F/A}{(F/A)_{st}} = \frac{(n_{H_2} + n_{CH_4})/n_{O_2}}{[(n_{H_2} + n_{CH_4})/n_{O_2}]_{st}} \quad (1)$$

where n_i is the mole number of each gas. The hydrogen mole fraction in the whole fuel is defined as

$$X_{H_2} = \frac{n_{H_2}}{n_{H_2} + n_{CH_4}} \quad (2)$$

Further, the dilution ratio of carbon dioxide to the whole mixture is represented as

$$Z_{CO_2} = \frac{n_{CO_2}}{n_{H_2} + n_{CH_4} + n_{CO_2} + n_{O_2} + n_{N_2}} \quad (3)$$

To evaluate the effect of hydrogen addition to the methane–air mixture diluted with carbon dioxide on the characteristics of the combustion behavior, the mole fraction of hydrogen in the whole fuel and the dilution ratio of carbon dioxide were varied in the ranges of 0–0.4 and 0–0.15, respectively. The initial pressure was 0.1 MPa, and the initial temperature of the mixture was approximately 283 K. Also, the equivalent ratio was set to 1 for all experimental conditions. Experiments were conducted three times for each data point. The constant-volume spherically expanding flame method has been widely used to laminar flame speed [33-35]. In this study, the edge of the flame front was detected using the Canny edge method in MATLAB. The flame radius is defined as $r = \sqrt{A/\pi}$, where A is the flame area scanned from the Schlieren image. The details of the image analysis are provided in Refs. [32, 36-38]. The flame speed, $S_n = dr/dt$, and the flame stretch rate, $K = d(\ln A)/dt = (1/A) dA/dt = (2/r) dr/dt$, were determined using the flame radius and time variation of the flame radius. Generally, there is a linear relationship between the stretched flame speed and the stretch rate [39, 40]. The stretched laminar flame speed S_n^0 is calculated as follows:

$$S_n^0 - S_n = L_b K \quad (4)$$

where L_b is the burned gas Markstein length [39, 40]. Finally, the unstretched laminar burning velocity S_u^0 is defined as:

$$S_u^0 = \frac{\rho_b}{\rho_u} S_n^0 = \frac{S_n^0}{\sigma} \quad (5)$$

where σ is the expansion ratio, and ρ_b and ρ_u are the burned and unburned gas densities, respectively. Furthermore, the combustion under the experimental conditions was computationally estimated by calculating a 1D planar flame using Chemkin-Pro [41] with the GRI Mech 3.0 reaction model [42]. The analysis conditions were set to be the same as the experimental conditions. The laminar burning velocity, flame structure, thermal diffusivity, reaction rate, and NOx concentration were analyzed by parsing the chemical reactions and, the experimental and analytical results were compared and discussed.

3. RESULTS AND DISCUSSION

3.1. Effect of CO₂ dilution and H₂ addition on the laminar burning velocity

The experimental and numerical S_u^0 values or results are compared in Fig. 1. The solid line represents the numerical data, whereas the symbols are the experimental data. The experimental data agree well with the numerical values. The results demonstrated that the laminar burning velocity increased with the hydrogen concentration, notwithstanding the decreasing tendency of the velocity with the increase in the carbon dioxide concentration. In particular, dilution with carbon dioxide significantly reduced the velocity. The laminar burning velocity decreases to less than half its original value when the dilution ratio of carbon dioxide is increased to $Z_{CO_2} = 0.1$, notwithstanding the addition of hydrogen. Due to the decrease in the laminar burning velocity, by diluting with carbon dioxide, the concentration of fuel-oxygen in the mixture is reduced, which reduces the reactivity of the fuel and oxygen. Furthermore, since carbon dioxide is a nonlinear triatomic molecule, it has a larger specific heat than those of hydrogen, methane, nitrogen, or oxygen and, thus, a lower thermal diffusivity. The relationship between the laminar burning velocity and the thermal diffusivity is represented as:

$$S_u^0 \approx \frac{\alpha}{\delta} \quad (6)$$

where α is the thermal diffusivity and δ is the laminar flame thickness. The calculated values of the thermal diffusivity as a function of the dilution ratio with carbon dioxide are shown in Fig. 2. The thermal diffusivity values increased with an increase in the hydrogen content and decreased with carbon dioxide dilution. This trend is like the aforementioned change in the velocity, suggesting that dilution with carbon dioxide decreases the thermal diffusivity; for example, when the dilution ratio is 0.1, the thermal diffusivity decreases by approximately 13%. Therefore, carbon dioxide absorbs the heat released by the oxidation reaction of the fuel, and the flame temperature does not increase as much as it would in the undiluted state. In contrast, the velocity increases with the addition of hydrogen. The reason for the increase in the laminar burning velocity is that hydrogen is a monatomic molecule with a low specific heat and a high thermal conductivity, which result in a large thermal diffusivity. As shown in Fig. 2, the thermal diffusivity increases with the addition of hydrogen, and when the addition rate was 0.4, the thermal diffusivity increased by approximately 5%. In addition, the reaction proceeds rapidly because the active H radical generated by the elementary reaction of hydrogen is more reactive than the CH₃ radical formed by the elementary reaction of methane–oxygen. The adiabatic flame temperature and the laminar flame thickness versus the dilution rate with CO₂ at different H₂ mole fractions are shown in Fig. 3. The laminar flame thickness is defined as:

$$\delta = \frac{T_{ad} - T_u}{(dT/dx)_{max}} \quad (7)$$

where T_{ad} is adiabatic flame temperature, T_u is unburned gas temperature and $(dT/dx)_{max}$ is the maximum value of temperature gradient. The adiabatic flame temperature decreased with the CO_2 dilution, notwithstanding an almost constant value with the hydrogen addition. Contrary to the results of adiabatic flame temperature, the increasing tendency of the flame thickness that describes the temperature profiles around the flame front location was observed with the increase in the carbon dioxide content, although the value slightly decreased with hydrogen addition. From these results, it can be seen that the laminar burning velocity increases with the addition of hydrogen according to equations (6) and (7), and conversely, the laminar burning velocity decreases when diluted with carbon dioxide.

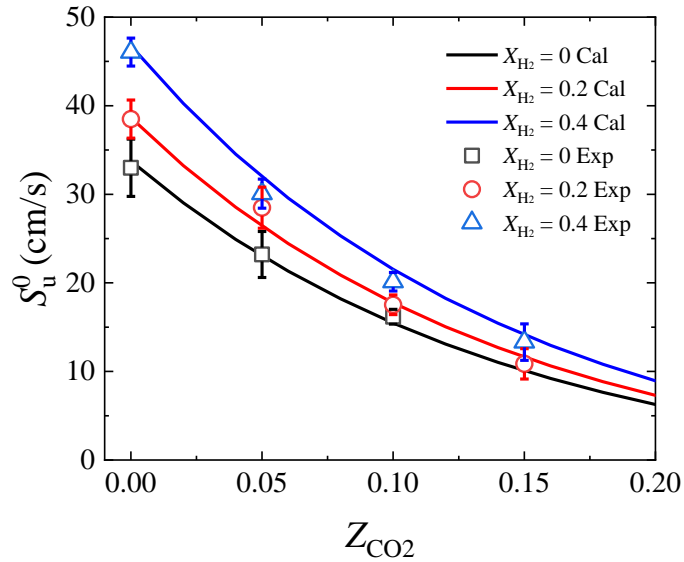


Figure 1. Unstretched laminar burning velocities at different carbon dioxide dilution and hydrogen addition.

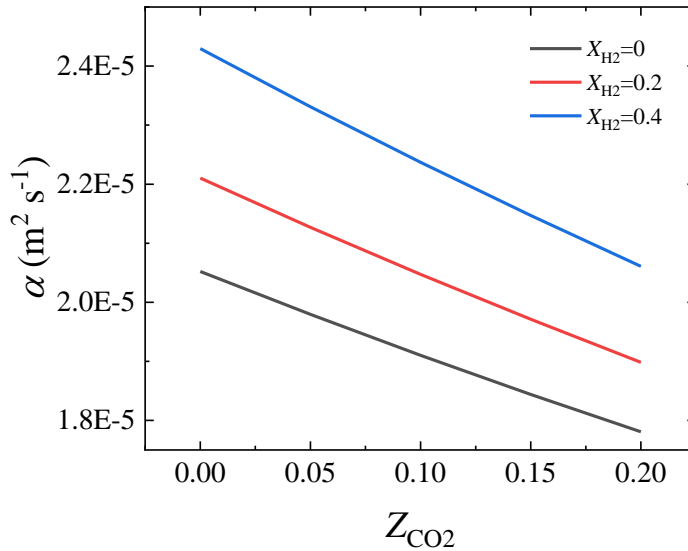


Figure 2. Thermal diffusivity at different carbon dioxide dilution and hydrogen addition.

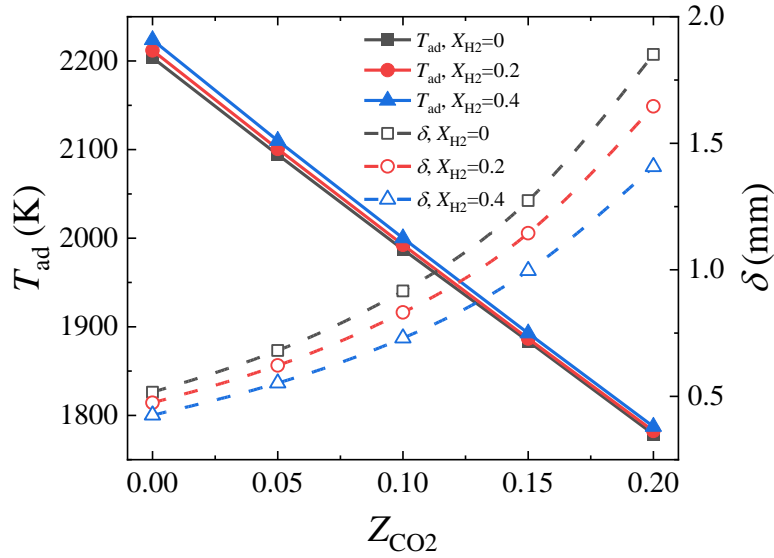
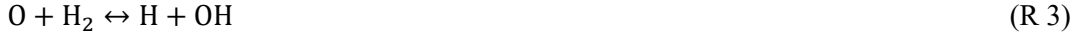


Figure 3. Adiabatic flame temperature and flame thickness at different carbon dioxide dilution and hydrogen addition.

3.2. Effect of CO₂ dilution and H₂ addition on the flame structure and reactions

A sensitivity analysis was performed to understand the effects of carbon dioxide dilution and hydrogen addition on the laminar burning velocity, flame temperature, and elementary reactions. The elementary reaction steps that mainly contributed to the increase in the combustion temperature of the hydrogen–methane–carbon dioxide–air mixtures are as follows [19]:



(R 3) and (R 38) are the most important chain branching reactions that produce many H, O, and OH radicals during combustion. In particular, (R 38) is the dominant reaction in the hydrogen combustion, and the sensitivity coefficient of (R 38) increases with increasing carbon dioxide addition and decreasing hydrogen addition. (R 97) and (R 119) play an essential role in the consumption of CH₃, the main active radical of methane, which is generated during the combustion of methane. Furthermore, (R 35), (R 52) and (R 99) are recombination reactions which consume active radicals and produce inactive substances. In particular, (R 99) converts the CO generated during the combustion of hydrocarbon fuels into CO₂, and its reaction rate is considerably lower than those of other reactions. Therefore, (R 99) is the rate-determining reaction that controls the oxidation of hydrocarbon fuels. The rates of these reactions increase as the concentration of radicals required for the reaction increases, thereby increasing the laminar burning velocity.

Figure 4 shows the flame temperature and mole fraction, x_i , of each active radical in the flame structure at different dilution ratios of carbon dioxide. The solid line in the graph shows the value without hydrogen addition, $X_{H_2} = 0$, and the dashed line shows the value with hydrogen addition, $X_{H_2} = 0.4$. As shown in Fig. 4, under all dilution conditions, the maximum concentrations of the active radicals, H, OH and O in the reaction zone increased with the addition of hydrogen. This is because the added hydrogen increases the production of the active H, OH and O radicals due to the promotion of the chain-branching reactions of hydrogen oxidation, as follows:



In other words, the addition of hydrogen increases the concentration of the active radicals in the reaction zone, thus increasing the collision and consequent reaction of radicals with each other. Sensitivity analysis of the temperature of the hydrogen-methane-carbon dioxide-air mixture shows that reactions (R 35), (R 38), (R 52) and (R 99) are dominant. Figure 5 shows the spatial distance, l , variation of the net reaction rate, ω , of (R 35), (R 38), (R 52) and (R 99) at different dilution ratios with carbon dioxide and the addition ratios of hydrogen. The results demonstrate that the net reaction rate tends to decrease with dilution using carbon dioxide, which is common in all reactions. This was attributed to the dilution, which reduced the concentration of gases and active radicals necessary for the reaction in the reaction zone. Conversely, the net reaction rate tends to increase with the addition of hydrogen. This is affected by the increase in the concentration of the activated radicals, H and OH in the reaction zone due to the addition of hydrogen. Figure 8 shows that the addition of hydrogen increases the reaction rates of all the reactions; in particular, the increase in the reaction rate of (R 38) is higher than those of the other reactions. (R 38) is faster than the recombination reactions that consume active radicals, which increase the concentration of active radicals in the reaction zone. In addition, the rate of (R 99) increases with the addition of hydrogen. Since (R 99) is the rate-determining reaction that controls the rate of oxidation of hydrocarbon fuels, an increase in the rate of (R 99) leads to an increase in the laminar burning velocity.

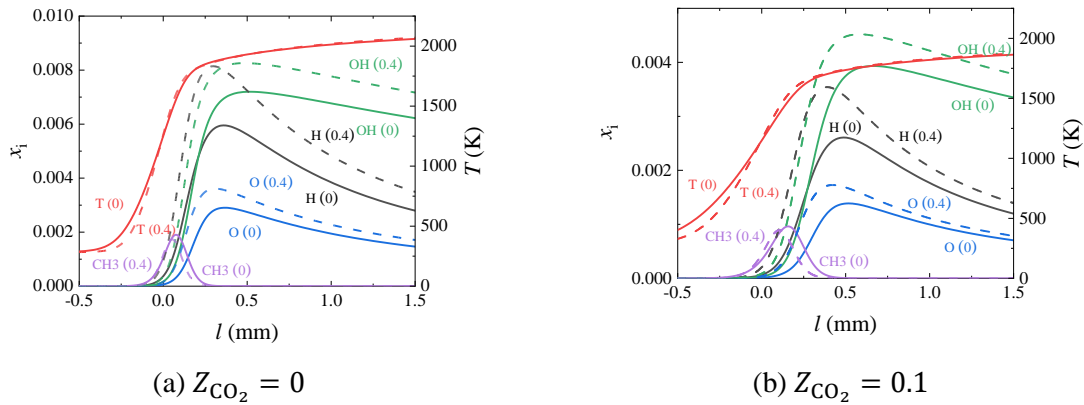
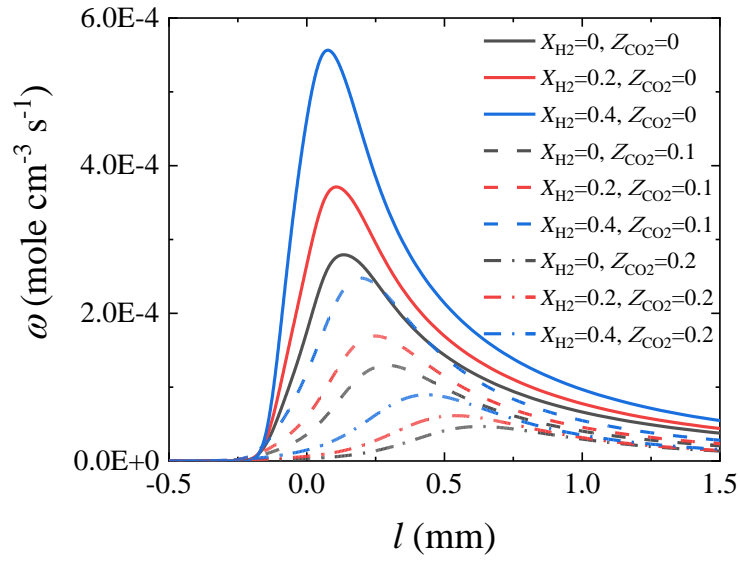
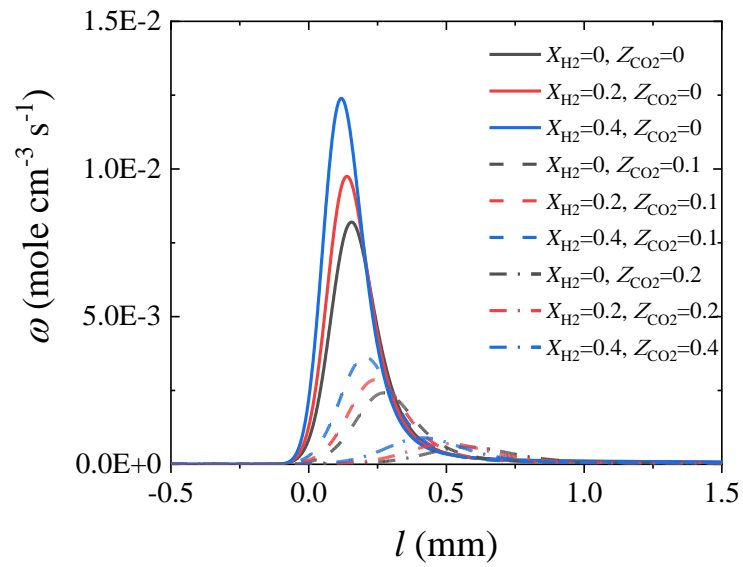


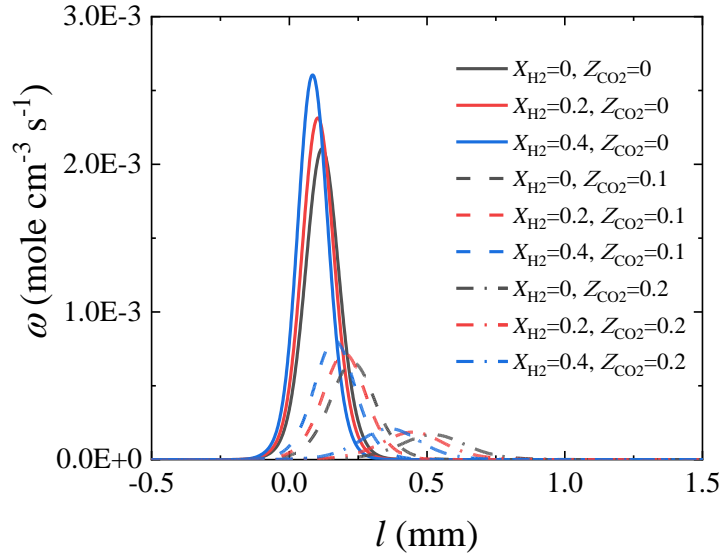
Figure 4. The mole fraction of active radicals in the flame structure



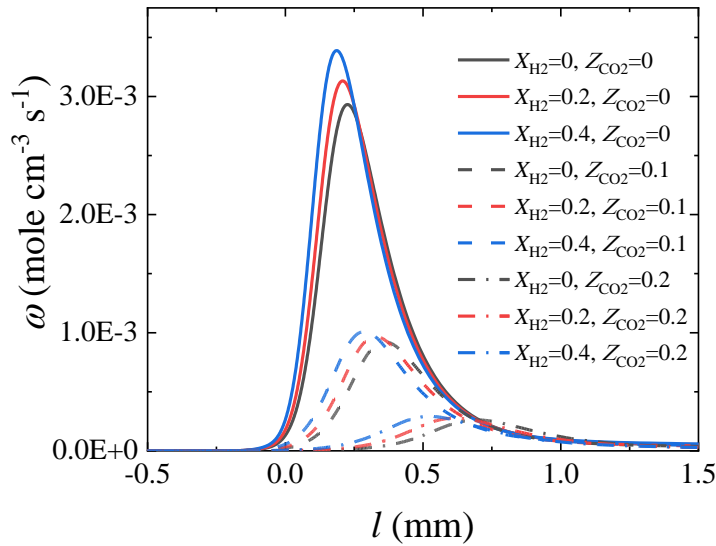
(a) (R 35): $\text{H} + \text{O}_2 + \text{H}_2\text{O} = \text{HO}_2 + \text{H}_2\text{O}$



(b) (R 38) : $\text{H} + \text{O}_2 = \text{O} + \text{OH}$



(c) (R 52): $\text{H} + \text{CH}_3(+\text{M}) = \text{CH}_4(+\text{M})$



(d) (R 99): $\text{OH} + \text{CO} = \text{H} + \text{CO}_2$

Figure 5. Spatial distance variation of net reaction rate

4. CONCLUDING REMARKS

In the present study, the combustion characteristics of a hydrogen–methane–carbon dioxide–air mixture at different hydrogen and carbon dioxide mole fractions were experimentally and numerically investigated. The values of the measured unstretched laminar burning velocities were compared with those obtained using a detailed chemical kinetic model. The experimental and numerical results agreed well. Overall, our results demonstrate the strong influence of hydrogen addition and carbon dioxide dilution on the laminar burning velocity of the mixtures. The velocity decreased as the mole fraction of carbon dioxide increased and conversely increased with the addition of hydrogen. These trends are

attributable to a change in the thermal diffusivity of the mixture, which increased with the addition of hydrogen and decreased with carbon dioxide dilution; the results are consistent with those of previous studies. With the carbon dioxide dilution, the flame thickness increased and the adiabatic flame temperature decreased. The rates of all the reactions increased with the dilution of carbon dioxide and increased with the addition of hydrogen. In particular, the rate of the dominant reaction, (R 38), which produces many active radicals, such as H, O and OH dramatically increases with hydrogen addition. The rate of (R 99), which is the rate-determining reaction of the hydrocarbon combustion, increased with the addition of hydrogen.

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