Laminar Burning Velocity of Stoichiometric CH₄/air Premixed Flames at High-Pressure and High-Temperature*

Yasuhiro OGAMI** and Hideaki KOBAYASHI**

Experimental and numerical studies on laminar burning velocities of stoichiometric CH₄/air flames were performed at high pressure and high temperature. A stoichiometric CH₄/air mixture was diluted by helium in order to restrain the intrinsic flame instabilities occurring at high pressure. Measurements of laminar burning velocity for burner-stabilized flames were conducted by a technique employing particle tracking velocimetry (PTV) and planar laser induced fluorescence for OH radical (OH-PLIF) simultaneously, which measures the instantaneous local burning velocity. Laminar burning velocities were determined by the average values of local burning velocities in the region where the Karlovitz number are sufficiently small, meaning that the effect of flame stretch and curvature can be neglected. Numerical simulations were also conducted using a one-dimensional premixed flame code. Detailed reaction mechanisms and the 4-step reduced mechanism were examined, and their results were compared with experimental results to investigate the feasibility of predicting the flame characteristics at high pressure and high temperature, based on the reaction mechanisms.

Key Words: Premixed Combustion, Burning Velocity, Chemical Reaction, High Pressure, High Temperature, Flame Stretch, PTV, OH-PLIF

1. Introduction

Laminar burning velocities, S_L , are one of the most important types of information for the analysis of combustion phenomena. Especially, for the design of high-load combustors such as gas turbines, S_L data at high pressure and high temperature are essential. In previous studies on CH₄/air premixed flames⁽¹⁾⁻⁽⁷⁾, S_L have been extensively investigated at atmospheric pressure and room temperature. However, the amount of reliable data on S_L in high-pressure and high-temperature environments is very limited mainly due to experimental difficulties.

One of the reasons for those difficulties is the intrinsic flame instability. At high pressure, Darrieus-Landau instability combined with the diffusive-thermal effect becomes significant, which makes measurement of S_L extremely difficult. This characteristic is common to all premixed flames. Therefore, conducting numerical simulation for one-dimensional flame is a better way to estimate S_L for various pressures and temperatures. In order to do this,

a reaction mechanism which is reliable even under such conditions is needed. Thus, verification of a suitable reaction mechanism using properly corrected experimental data should be performed, although existing experimental data is limited to certain mixtures in certain ranges of pressure and temperature.

Another difficulty in S_L measurements is the exclusion of flame stretch. Although S_L is the propagating speed of a non-stretched flame, the flames produced using various experimental techniques such as a spherical bomb technique or a twin-flame technique are affected by the flame stretch, and their propagating speed must be different from S_L . Therefore, for the extraction of S_L , some extrapolations to a non-stretched flame are necessary. However, the difference between the ways in which extrapolation is done by various researchers often results in different S_L data despite the use of the same experimental technique. Accordingly, another method by which S_L can be obtained directly from experimental data without any extrapolation is preferable.

Recently, to solve these two problems, measurements of S_L for a stoichiometric H₂/O₂/He was performed using a new technique which can determine the local burning velocities of burner-stabilized premixed flames based on

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** Institute of Fluid Science, Tohoku University, 2-1-1 Katahira, Aoba-ku, Sendai, Miyagi 980-8577, Japan.
E-mail: ogami@flame.ifs.tohoku.ac.jp

particle tracking velocimetry (PTV) and planer laser induced fluorescence for the OH radical (OH-PLIF) at elevated pressure and elevated temperature⁽⁸⁾, which is an extension of the experimental method employed in our previous studies^{(9),(10)}. This experimental method has two advantages. One is helium dilution of the mixture, which makes the Lewis numbers of fuel and oxygen larger than unity. With the increase of Lewis numbers, Darrieus-Landau instabilities are lessened by the diffusive-thermal effect⁽¹¹⁾. Actually, due to the helium dilution, stoichiometric $H_2/O_2/He$ flames become stable over a wide range of pressure from 0.1 to 1.0 MPa and mixture temperature from 300 to 500 K⁽⁹⁾. The same strategy has been used in other studies^{(12),(13)}. Another advantage is that this new experimental technique⁽⁸⁾ provides S_L information directly from experimental data, so that no extrapolation to the non-stretched flame is necessary. Another direct measuring method of measurement exists⁽⁵⁾, but only our experimental method can treat a stable flame. At elevated pressure, because flame instabilities are significant, methods employing with continuous observation of the flame front are preferable.

In this study, using the same strategy, S_L for a stoichiometric $CH_4/air/He$ mixture were measured at elevated pressure and elevated temperature. A stoichiometric mixture is suitable for the verification of the reaction mechanisms, because the effect of the radiation heat loss is small even at high pressure. Numerical simulations using detailed reaction mechanisms and a 4-step reduced mechanism were also conducted. The experimental and numerical results are herein compared, and the applicability of reaction mechanisms for various conditions of pressure and temperature are discussed.

2. Experimental and Numerical Methods

2.1 Experimental apparatus

Figure 1 shows a schematic diagram of the experimental apparatus. A high-pressure combustion chamber with an inner diameter of 250 mm and a height of 1050 mm was employed in this study. Compressed air from the air tank was supplied continuously from the bottom of the combustion chamber, and the pressure in the combustion chamber was kept constant by adjusting the exhaust valve. A nozzle-type burner was installed in the combustion chamber. In order to reduce the effects of flame stretch and curvature in the longitudinal direction of the burner, slot-type nozzle burners were used. Depending on the experimental conditions, three sizes of burner (outlet dimensions of 8 mm × 15 mm, 4 mm × 15 mm, and 4 mm × 10 mm) were employed. Methane, air and helium gases were supplied to the burner from bombs. A heater was installed in the combustion chamber, and air and helium were heated before mixing with methane. The actual temperature of the mixture was measured by a thermocou-

Fig. 1 Schematic diagram of experimental setup

ple installed close to the burner outlet (5 cm upstream of the burner outlet).

For PTV measurement, a double-pulsed Nd-YAG laser (Continuum, Minilite) was used. The Nd-YAG laser beam was transformed into a vertical sheet with a thickness of less than 100 μm by conical and cylindrical lenses. SiO_2 particles with specific gravity and a mean diameter of 2.0 and 2.0 μm , respectively, were added to the mixture as Mie scattering particles, and their images were obtained by a CCD camera (Kodak, Megapulus Es1.0; 1008 × 1014 pixels). A dye laser (Spectron, SL-4000G), pumped by another Nd-YAG laser (Continuum, Surelite) and fired in the center of double-pulse Nd-YAG laser outputs, was used for OH-PLIF. The dye laser output was also transformed into a sheet and superposed onto the Nd-YAG laser sheet. It excited a blended branch of $Q_1(9)$ and $Q_2(8)$ of the OH (1,0) band. OH-PLIF images were taken from the opposite side of the CCD camera for PTV using an intensified CCD camera (Princeton Instruments, ICCD576/RB-T, 576 × 384 pixels).

The measurements of S_L were performed in the range of pressure, P , of 0.1 to 1.0 MPa, and in the range of mixture temperature at the burner outlet, T_0 , of 298 to 600 K. The accuracy of temperature throughout the experiments was ± 3 K.

2.2 Numerical method

Numerical simulations of one-dimensional premixed flames were conducted using PREMIX⁽¹⁴⁾. The thermochemical data and transport properties were evaluated using CHEMKIN-II⁽¹⁵⁾. GRI-Mech 3.0⁽¹⁶⁾ and GRI-Mech 2.11⁽¹⁷⁾ were considered as detailed reaction mechanisms. In addition to detailed mechanisms, the 4-step reduced mechanism proposed by Peters⁽¹⁸⁾ was also used, because reduced mechanisms are very effective for computational time efficiency and should be tested as to their feasibility at high pressure and high temperature. C1-chemistry by Warnatz⁽¹⁹⁾, which is the starting mechanism of the 4-step mechanism, was also employed.

Fig. 2 OH-PLIF image of the flame at 1.0 MPa: (a) $\text{CH}_4/\text{air}/\text{He}$ ($\phi = 1.0$, $T_0 = 400 \text{ K}$); (b) CH_4/air ($T_0 = 298 \text{ K}$, $\phi = 1.0$, $\text{He} = 10 \text{ vol.}\%$)

None of the reaction mechanisms contain information about the Chaperon efficiencies of helium. Therefore, we used the Chaperon efficiencies of argon for helium because of the thermochemical similarities of these species, following the method of Kwon et al.⁽¹³⁾ Although this seems to be rough treatment, it was confirmed that the difference of the Chaperon efficiency of helium produced only slight change of numerical results in the conditions tested. As mentioned later, the amount of helium was very small in this study. Moreover, the Chaperon efficiencies of helium are relatively smaller than those of other species. Therefore, such use of the Chaperon efficiencies does not affect numerical results.

The effect of radiative heat loss was not considered, because it is very small in the case of a stoichiometric CH_4/air mixture. Actually, the numerical results using an optically-thin model showed small differences of less than 1% with respect to those of the adiabatic model in all of the conditions tested in this study.

3. Results and Discussion

3.1 Restraint of intrinsic flame instability

As mentioned above, the intrinsic flame instability, i.e., Darrieus-Landau instability combined with diffusive-thermal instability, becomes significant at high pressure. In the case of stoichiometric CH_4/air premixed flames, flame instability was observed from 0.4 MPa. In this study, in order to restrain flame instabilities, a stoichiometric CH_4/air mixture was diluted by helium to a helium dilution ratio of 10% in volume. This dilution makes the Lewis number of the mixture, $L_e (= \alpha/D$, α : thermal diffusivity, D : diffusion coefficient), more than unity, and Darrieus-Landau instability is lessened by the diffusive-thermal effect⁽¹⁰⁾. When the helium dilution ratio is zero, i.e., a stoichiometric CH_4/air mixture, the L_e of methane and that of oxygen are 1.0 and 1.1, respectively. However,

at the 10% helium dilution ratio, they are increased to 1.2 and 1.3, respectively, so that Darrieus-Landau instability is restrained by the diffusive-thermal effect. Actually, as shown in Fig. 2 (a), a $\text{CH}_4/\text{air}/\text{He}$ flame was observed to be stable even at 1.0 MPa, although a CH_4/air flame was strongly distorted by the flame instability at this pressure as seen in Fig. 2 (b). Measurements of S_L at high pressure and high temperature were performed using this mixture; i.e., a stoichiometric $\text{CH}_4/\text{air}/\text{He}$ mixture of the 10% helium dilution ratio.

3.2 Determination of laminar burning velocity

A set of binarized particle images collected by the CCD camera was analyzed using VISIFLOW (AEA Technology), and a map of instantaneous velocity vectors over the cross section of the flame was generated (Fig. 3 (a)). Then, from this map, the local streamlines were calculated by interpolation. Along each streamline, the point where the magnitude of a velocity vector changes abruptly was determined. The OH-PLIF image was also taken by the ICCD camera at the same time as PTV (Fig. 3 (b)), and the flame front was determined from the OH-PLIF image at the point where LIF intensity begin to increase, and was fitted to the polynomial curve and superposed on the streamlines (Fig. 4). At the intersection of the calculated local streamline and the flame front, after calculating the angle between the tangential line of the flame front and the streamline, θ_1 , the local burning velocity, S_r , was obtained using the following equation: $S_r = U_r \sin \theta_1$, where U_r is the magnitude of the local velocity vector at the front edge of the preheating zone.

Validity of the experimental method used in this study can be confirmed as follows. Figure 5 shows distributions of S_r and Karlovitz number estimated from the local velocity gradient, K_a , ($= \varepsilon \alpha / S_L^2$, α : thermal diffusivity, ε : stretch rate), for a stoichiometric CH_4/air premixed flame at 0.1 MPa and 298 K. This result shows large values of

Fig. 3 Velocity vectors and OH-PLIF image used for the measurement of local burning velocities (CH_4/air , $P = 0.1 \text{ MPa}$, $T_0 = 298 \text{ K}$, $\phi = 1.0$): (a) Velocity vectors; (b) OH-PLIF image

Fig. 4 Flow stream lines and flame front

S_r near the burner lip. In this region, K_a are also large, so that an increase of S_r is caused by the flame stretch due to the boundary layer. Between the burner lip and flame tip, however, there is a region where S_r values are nearly constant. In this region, K_a are $O(10^{-3})$, and flame curvature and stretch are minimum. According to Hassan et al.⁽⁶⁾, Markstein number in this condition is 0.95, so that the difference of S_r from S_L due to the flame stretch is estimated to be less than 1% and thus the flame is considered to be non-stretched flame. That is why the mean value of S_r in this region is considered to be S_L . In all experimental conditions, similar magnitude of K_a were obtained, that is, the effect of flame stretch was sufficiently small.

To test this experimental technique, S_L measurements of CH_4/air premixed flames for various equivalence ratios were performed at atmospheric pressure and room temperature, and comparison of experimental data with

Fig. 5 Distributions of local burning velocity, S_r , and Karlovitz number, K_a (CH_4/air ; $P = 0.1 \text{ MPa}$, $T_0 = 298 \text{ K}$, $\phi = 1.0$)

Fig. 6 Relationship between laminar burning velocity, S_L , and equivalence ratio, ϕ (CH_4/air ; $\phi = 1.0$, $P = 0.1 \text{ MPa}$, $T_0 = 298 \text{ K}$)

other experimental data was conducted. Figure 6 shows the experimental results of S_L with other recent data by Vagelopoulos et al.⁽⁴⁾, Hassan et al.⁽⁶⁾ and Rozenchan et al.⁽⁷⁾, and numerical results using GRI-Mech 3.0⁽¹⁶⁾ and GRI-Mech 2.11⁽¹⁷⁾. Error bars indicate the variation of

Fig. 7 Relationship between laminar burning velocity, S_L , and ambient pressure, P (CH_4/air ; $\phi = 1.0$, $T_0 = 298 \text{ K}$)

S_r in the region where S_L are determined. Experimental data of the present work are in good agreement with those of Hassan et al.⁽⁶⁾ and Rozenchan et al.⁽⁷⁾ Data by Vagelopoulos et al.⁽⁴⁾ are in good agreement with numerical results, although experimental data of the present work, Hassan et al.⁽⁶⁾ and Rozenchan et al.⁽⁷⁾ differ at low equivalence ratios. This is because the optimization of GRI-Mech 3.0⁽¹⁶⁾ and GRI-Mech 2.11⁽¹⁷⁾ were conducted using the data by Vagelopoulos et al.⁽⁴⁾

3.3 Laminar burning velocity for CH_4/air premixed flame at high pressure and room temperature

Before the measurements of S_L for $\text{CH}_4/\text{air}/\text{He}$ mixtures at elevated pressure and elevated temperature, S_L for a stoichiometric CH_4/air mixture at high pressure and room temperature were performed. As mentioned above, a stoichiometric CH_4/air mixture is unstable at pressure higher than 0.4 MPa due to Darrieus-Landau instability, so that data are limited up to 0.3 MPa. Figure 7 shows the pressure dependency of S_L for stoichiometric CH_4/air premixed flames. Recent experimental data by Egolfopoulos et al.⁽³⁾, Hassan et al.⁽⁶⁾ and Rozenchan et al.⁽⁷⁾, and numerical results using GRI-Mech 3.0⁽¹⁶⁾ and GRI-Mech 2.11⁽¹⁷⁾ are also plotted in Fig. 7. Even at high pressure, experimental results of the present work are in good agreement with those of Hassan et al.⁽⁶⁾, and marginally in good agreement with those of Rozenchan et al.⁽⁷⁾ and Egolfopoulos et al.⁽³⁾ The experimental data of present work also marginally agree with numerical results using GRI-Mech 3.0⁽¹⁶⁾ in this pressure range at room temperature.

3.4 Laminar burning velocity for $\text{CH}_4/\text{air}/\text{He}$ premixed flame for various conditions

Figure 8 (a), (b), (c) and (d) show experimental data and numerical results using GRI-Mech 3.0⁽¹⁶⁾, GRI-Mech 2.11⁽¹⁷⁾, the 4-step mechanism by Peters⁽¹⁸⁾, and C1-chemistry by Warnatz⁽¹⁹⁾ for $\text{CH}_4/\text{air}/\text{He}$ premixed flames at T_0 of 298, 400, 500 and 600 K, respectively. All experimental results are in good agreement with numerical

Fig. 8 Relationship between laminar burning velocity, S_L and ambient pressure, P ($\text{CH}_4/\text{air}/\text{He}$, $\phi = 1.0$, $\text{He} = 10\%$ in vol.): (a) $T_0 = 298 \text{ K}$; (b) $T_0 = 400 \text{ K}$; (c) $T_0 = 500 \text{ K}$; (d) $T_0 = 600 \text{ K}$

results using GRI-Mech 3.0⁽¹⁶⁾, GRI-Mech 2.11⁽¹⁷⁾, and C1-chemistry by Warnatz⁽¹⁹⁾ at atmospheric pressure in all conditions of T_0 . Numerical results using GRI-Mech 3.0⁽¹⁶⁾ are also marginally in good agreement with experimental data at T_0 of 298 and 400 K, even at high pressure. However, the differences between numerical results using

Fig. 9 Sensitivity coefficients ($\text{CH}_4/\text{air}/\text{He}$, $\phi = 1.0$): (a) $T_0 = 298$ K; (b) $T_0 = 600$ K

GRI-Mech 3.0⁽¹⁶⁾ and our experimental results become significant at high pressure as T_0 increases. In contrast, numerical results using GRI-Mech 2.11⁽¹⁷⁾ are in good agreement with experimental data at T_0 of 500 and 600 K, although they are larger at 298 and 400 K. It is apparent that both GRI-Mech 3.0⁽¹⁷⁾ and GRI-Mech 2.11⁽¹⁶⁾ have deficiencies in the pressure dependency, i.e., the tendency of S_L to decrease as pressure increases. This is because GRI-Mech 3.0⁽¹⁷⁾ and GRI-Mech 2.11⁽¹⁶⁾ are optimized using a small number of S_L data at elevated temperature, and thus further development using a large number of experimental data at elevated pressure and elevated temperature seems to be necessary. Numerical results using the 4-step mechanism of Peters⁽¹⁸⁾ have large disagreement in the pressure dependency of S_L at all T_0 . Numerical results using C1-chemistry by Warnatz⁽¹⁹⁾, which is the starting mechanism of the 4-step mechanism of Peters⁽¹⁷⁾, also have large differences in the S_L pressure dependency. This is one of the reasons why the results using the 4-step mechanism of Peters⁽¹⁷⁾ have large differences in pressure dependency. Furthermore, quasi steady-state assumption performed in the reduction of the 4-step reaction mechanism does not seem to be adequate, especially at high pressure and high temperature, leading to the disagreement of

numerical results when the 4-step mechanism of Peters⁽¹⁷⁾ is used.

3.5 Sensitivity analysis

For understanding which reaction has a large effect on the flame propagation in the $\text{CH}_4/\text{air}/\text{He}$ system at high pressure and high temperature, sensitivity analysis was performed using GRI-Mech 3.0⁽¹⁶⁾. Figures 9(a) and (b) show numerical results of sensitivity analysis with respect to S_L in the $\text{CH}_4/\text{air}/\text{He}$ system at 0.1, 0.5 and 1.0 MPa for $T_0 = 298$ and 600 K, respectively. In these figures, results for 26 reactions which have large sensitivity are presented. Because of the small amount of additional helium, results of sensitivity analysis are almost the same as those of the CH_4/air system in the conditions tested in this study. Although small differences of sensitivities exist between $T_0 = 298$ and 600 K, the reactions which have large sensitivities are similar. Chain propagating and branching reactions, which exhibit large positive sensitivities, are $\text{H} + \text{O}_2 = \text{O} + \text{OH}$, $\text{OH} + \text{CH}_3 = \text{CH}_2(\text{S}) + \text{H}_2\text{O}$, $\text{OH} + \text{CO} = \text{H} + \text{CO}_2$ and $\text{HO}_2 + \text{CH}_3 = \text{OH} + \text{CH}_3\text{O}$. Recombination reactions which have large negative sensitivity are $\text{H} + \text{O}_2 + \text{H}_2\text{O} = \text{HO}_2 + \text{H}_2\text{O}$ and $\text{H} + \text{CH}_3 + \text{M} = \text{CH}_4 + \text{M}$. As pressure increases, sensitivities of the reactions $\text{H} + \text{O}_2 = \text{O} + \text{OH}$, $\text{HO}_2 + \text{CH}_3 = \text{OH} + \text{CH}_3\text{O}$ and $\text{H} +$

$\text{CH}_3 + \text{M} = \text{CH}_4 + \text{M}$ become large, i.e., these reactions have a large effect on the S_L pressure dependency. This tendency for these three reactions with respect to pressure is the same even at high temperature, meaning that they are reactions which have a strong effect on determining S_L at high pressure and high temperature.

4. Conclusions

Measurement of laminar burning velocities for preheated $\text{CH}_4/\text{air}/\text{He}$ stoichiometric mixtures stabilized by a slot-type nozzle burner in a high-pressure chamber were conducted based on a technique using particle tracking velocimetry and planar laser induced fluorescence. Numerical analyses were also performed using detailed reaction mechanisms and a reduced mechanism in order to test the validity of the existing reaction mechanisms for the flame at elevated pressure and temperature.

The following results were obtained:

1. The intrinsic flame instability at high pressure was restrained due to the increase in Lewis number by helium dilution, and thus measurements of laminar burning velocities were successfully performed up to a pressure of 1.0 MPa and a mixture temperature of 600 K for stable flames.
2. Numerical results of S_L using GRI-Mech 3.0⁽¹⁶⁾, GRI-Mech 2.11⁽¹⁷⁾ and C1-chemistry by Warnatz⁽¹⁹⁾ are in good agreement with experimental data at atmospheric pressure even at high temperature, although the discrepancy with experimental results becomes significant with the rise in pressure depending on the reaction mechanisms.
3. Numerical results of S_L using the 4-step mechanism of Peters⁽¹⁷⁾ are quite different from the experimental data according to the pressure dependency of S_L at all mixture temperatures.

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