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A STUDY ON TURBULENT BURNING VELOCITY OF METHANE MIXTURES WITH HYDROGEN ADDITION

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ABSTRACT The purpose of this paper is to investigate the combustion properties of methane, which is the main component of natural gas and biogas and to find out the methods for improving the turbulent combustion performance. To realize this, a new combustion technique, hydrogen addition to the methane mixture, has been proposed and studied quantitatively in detail. Consequently, it is shown that by adding only a small amount of hydrogen to the lean methane mixture, the turbulent burning velocity is substantially increased. Furthermore these mechanisms are discussed in view of the change in the local burning velocity due to the flame stretch. As a result, it is concluded that the change in the Markstein number among the mixtures, mainly because of the change in the Lewis number by hydrogen addition, accounts for the change in the turbulent combustion properties.

Keywords: Internal Combustion Engine, Premixed Lean Combustion, Hydrogen Addition, Flame Stretch

1. INTRODUCTION

Recently there are many problems to be urgently tackle in worldwide scale. One of such problems is environmental issue such as global warming caused by increasing carbon dioxide. The other is energy problem such as the exhaustion of fossil fuels. These social circumstances force the internal combustion engine to take some realistic measures urgently. For this purpose, a number of alternative fuels and new combustion techniques have been investigated. As one of the promising alternative fuels, methane, which is the main component of natural gas and biogas, has attracted considerable attention. Therefore, the clarification of the combustion properties of methane mixture is important from the application point of view.

Meanwhile, with regard to the combustion techniques for internal-combustion engines, one of the most effective ways to improve the thermal efficiency and to cut down the pollutant emission is the lean-burn technique. Although this method has already been in practical use, there are many technical problems, such as the substantial decrease in the burning velocity and the large increase in the misfire probability in the lean mixture region. To solve these problems, a number of methods have been proposed so far, yet the problems are not settled completely. To improve the lean-burn technique, the authors have proposed a new method, hydrogen addition to hydrocarbon mixtures, and shown that the method work effectively on the improvement of the combustion performance for lean hydrocarbon mixtures, particularly for methane mixtures [1].

The purpose of this paper is to clarify the general properties of methane flames and to study in detail the

effects of hydrogen addition to methane mixtures for investigation of the feasibility to utilize methane in lean burn engines. As a result, it is found that the effects of hydrogen addition can be explained by the change in the local dependence of combustion properties on the flame stretch, that is, the change in Markstein number.

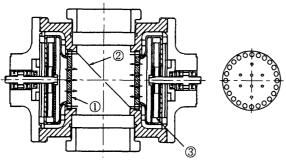
2. EXPERIMENT

2.1 Combustion Chamber

The combustion chamber used in this study is a nearly spherical vessel with a mean inner diameter of about 100mm. It is fixed with four transparent windows of 92mm diameter at four sides for flame observation, and two perforated plates of 92mm diameter at two opposite sides. The cross-sectional view of the combustion chamber observed from the transparent window is shown in Fig.1. Behind each perforated plate, a fan is equipped to mix gases and generate approximately isotropic and homogeneous turbulence in the central region of the chamber. The characteristics of turbulence in the chamber have been obtained by the two-point correlation method using LDV.

2.2 Measurement of burning velocity

In this study, both laminar and turbulent burning velocities were determined from measurements of pressure rise. Such a burning velocity can be related to the mass burning rate [2]. The laminar burning velocity, S_{L0} , was measured by the following procedure. The stretched laminar burning velocity, S'_{L0} , was calculated by the



1: Perforated Plate; 2:Ignition Plug; 3:Fan

Figure 1 Combustion Chamber

Table 1 Properties of mixtures

Mixture	δ	Φ	T_{ad}	S_{L0}	D_{th}	Le
		:	^{.і.} [К]эт	[cm/s]	10 ⁻⁵ [m ² /s]	
M06H00S20		0.6	221Ï	19.61	2.207	0.960
M07H00S20		0.7	2227	19.56	2.208	0.961
M08H00S20	0	0.8	2296	20.14	2.209	0.960
M09H00S20		0.9	2348	19.92	2.210	0.960
M10H00S20		1.0	2404	19.77	2.211	
M11H00S20		1.1	2416	19.60	2.212	1.093
M06H01S20		0.6	2184	19.54	2.263	0.284
M07H01S20		0.7	2217	19.76	2.266	0.285
M08H01S20	0.1	0.8	2278	19.92	2.269	0.285
M09H01S20		0.9	2323	19.96	2.283	0.286
M10H01S20		1.0	2389	20.26	2.277	
M11H01S20		1.1	2396	20.42	2.284	1.121
M06H02S20		0.6	2150	19.87	2.324	0.290
M07H02S20		0.7	2193	19.64	2.329	0.290
M08H02S20	0.2	0.8	2235	20.12	2.333	0.291
M09H02S20		0.9	2296	19.76	2.339	0.292
M10H02S20		1.0	2362	19.73	2.349	
M11H02S20		1.1	2377	20.16	2.364	1.150

pressure history of combustion according to Lewis's method [2]. One test to calculate S'LO was carried out using the pressure increase rate averaged over a range of gauge pressure between 0.05[atm] and 0.150[atm], where the variations of the calculated burning velocity with increasing flame radius (that is, with decreasing flame stretch) were less than 5 %. This is because the laminar flames have relatively large radius (that is, small stretch) over the range of pressure used for calculation. The values of S'_{L0} were finally obtained by averaging the results of 10 tests at each condition. In this case, the experimental uncertainties of S'LO due to the averaging process were less than 5%. So the laminar burning velocity, S_{L0}, were approximated directly by the values, S'_{L0} without correction, that is, it was assumed that the flame stretch effects on a laminar spherical flame were negligible under the current experimental circumstances. The turbulent burning velocity, approximated S_T , is by the relation [3], $S_T/S_{L0}=(dp/dt)_T/(dp/dt)_L$, where dp/dt is the pressure increase rate, and subscripts T and L designate turbulent and laminar combustion, respectively. The experimental

uncertainties for measuring the turbulent burning velocities were less than 5%.

2.3 Mixture properties

In this study, mixtures were mainly identified by two parameters; the total fuel/air equivalence ratio, Φ , and the volume fraction of the added hydrogen in the whole fuel, δ . The compositions can be expressed as

$$(1 - \delta)CH_4 + \delta H_2 + X_0O_2 + X_1N_2$$

In this case, Φ is calculated by $\Phi = (2 - 3/2 \delta)/X_0$

Where X₀ and X₁ are the mole numbers of oxygen and nitrogen, respectively. The burning velocity of a flame can be controlled independently of the equivalence ratio by changing the dilution with nitrogen (dilution = $X_0/(X_0+X_1)$). Table 1 shows that the properties of the mixtures with S_{L0} =20cm/s over a range of Φ between 0.6 and 1.1 for each $\delta = 0, 0.1, 0.2$. Where T_{ad} is the adiabatic flame temperature calculated by equilibrium calculation, Dth is the thermal diffusivity, Le is the Lewis number based on the molecular diffusion coefficient of the deficient reactants. Le is the Lewis number based on the molecular diffusion coefficient of the deficient reactants. And Lewis numbers for mixtures with hydrogen were calculated using the diffusion coefficient of hydrogen, because for these mixtures, the high mass diffusion of hydrogen is expected to play an important role in characteristics of a stretched flame. All thermodynamic and transport properties are evaluated by CHEMKIN[4][5].

3. RESULTS AND DISCUSSION

3.1 The effects of equivalence ratio on the turbulent combustion performance

Firstly the effects of equivalence ratio on the turbulent burning velocity have been investigated by using the mixtures tabulated in Table1. Figure 2 shows the experimental results on the turbulent burning velocity, S_T , plotted against the turbulence intensity, u^\prime , where both are non-dimensionalized by the laminar burning velocity, S_{L0} . From Fig.2, it is found that the S_T/S_{L0} increases with decreasing Φ at a certain u^\prime/S_{L0} for each $\delta{=}0,\,0.1,\,0.2.$ This trend holds true for all CH_4/H_2 fuel mixtures ranging from $\delta{=}0$ to $\delta{=}1$, though those figures are not included due to limitation of space. From this result, it can be concluded that CH_4/H_2 fuel is suitable for lean-burn technique considering the improvement of the turbulent combustion performance in the lean mixture region regardless of the ratio of CH_4 to H_2 .

3.2 The quantitative effects of hydrogen addition on the turbulent combustion performance

Secondly, the quantitative effects of hydrogen addition on the turbulent burning velocity have been experimentally examined by using the same mixtures as those ranging from δ =0 to 1.0 in the last paragraph. Figure 3 shows that the variations of S_T/S_{L0} with u^*/S_{L0} over a range of δ between 0 and 1.0 for each equivalence ratio, Φ =0.6, 0.8, 1.0. From

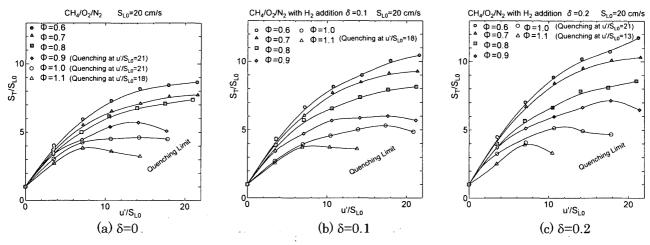


Figure 2 The effects of equivalence ratio on the turbulent burning velocities

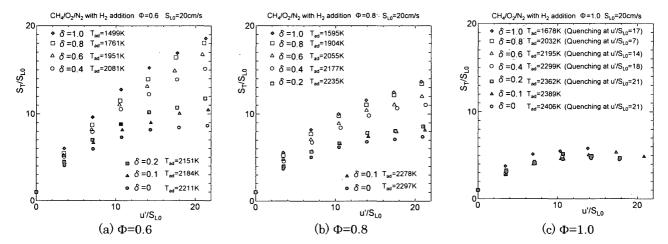


Figure 3 The quantitative effects of hydrogen addition on the turbulent velocities

the figures, it is found that with the addition of hydrogen, the turbulent burning velocities become correspondingly higher in the lean mixture region (Φ =0.6). The differences among the mixtures decrease, as the equivalence ratio approaches unity (Φ =0.8). For the stoichiometric mixtures (Φ =1.0), there is no appreciable change in S_T/S_{L0} with δ . In addition, as for the quenching limit of the stoichiometric mixtures, u'/ S_{L0} is about 10 for a range of δ , between 0.5 and 0.9, while u'/ S_{L0} is about 20 for a range of δ , between 0.1 and 0.4. Namely, the quenching limit decreases, as more hydrogen is added near stoichiometric condition.

To summarize the above, the hydrogen addition to methane mixtures improves the turbulent combustion performance with amounts of hydrogen in the lean mixture region, to be sure, yet it could have no effect or, in some cases, negative effect on the turbulent combustion performance near stoichiometric conditions. In other words, it might be said that small amounts of hydrogen additions are safer in consideration of the balance between the increase in the combustion performance and the small fluctuation of the operating condition due to the perturbation of mixture compositions.

3.3 The effects of dilution on the turbulent combustion performance

Thirdly, the effects of dilution with nitrogen on the turbulent combustion performance are examined, taking account of the reduction of the pollutant emission through the temperature control. Figure 4 shows the experimental results on S_T/S_{L0} with u^\prime/S_{L0} by using mixtures having a variety of S_{L0} for each $\delta{=}0,\,0.1,\,0.2$ at the equivalence ratio, $\Phi=0.6,\,0.8,\,1.0$, respectively. From the figures, it is found that relationships between u^\prime/S_{L0} and S_T/S_{L0} are almost identical for each δ at a specified $\Phi_{,}$ independent of the laminar combustion properties (e.g. burning velocity, S_{L0} , adiabatic flame temperature, T_{ad} , or laminar flame thickness estimated by D_{th}/S_{L0}). This indicates that the discussion for the mixtures having $S_{L0}{=}20\text{cm/s}$ in the previous paragraphs can hold true for all $CH_4/H_2/O_2/N_2$ mixtures regardless of dilution. That is,

- · For the lean mixtures S_T/S_{L0} increases with δ at a certain u^2/S_{L0} .
- · The differences in S_T/S_{L0} with δ among the mixtures decrease as Φ approaches unity.
- \cdot For the rich mixtures, S_T/S_{L0} does not change, or, in some cases, decreases with δ at a certain $u'/S_{L0}.$

Thus, again these results suggest that the hydrogen addition to lean mixtures can improve the combustion performance, independent of the dilution with nitrogen.

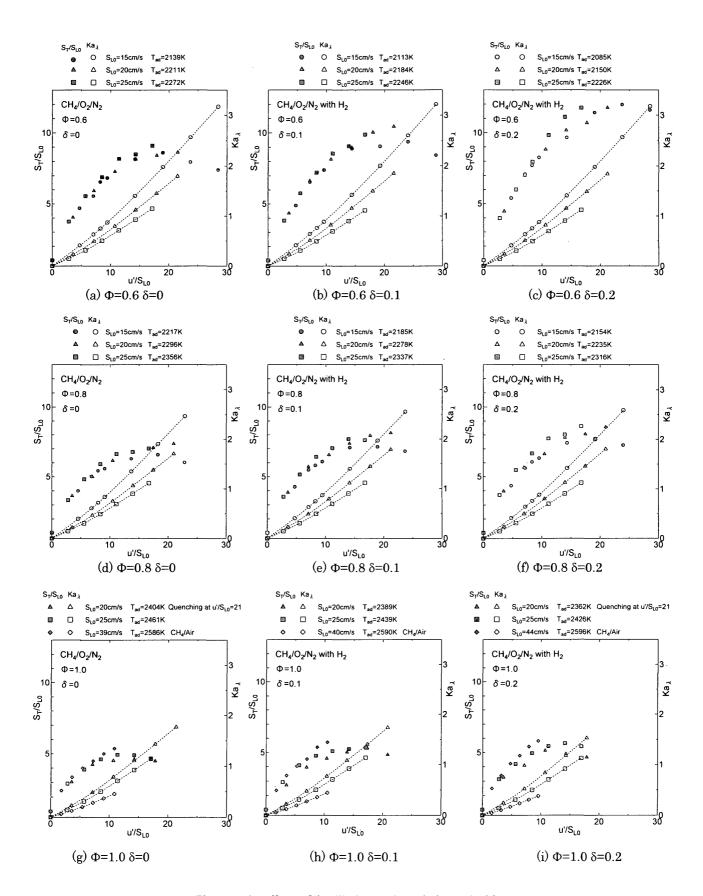
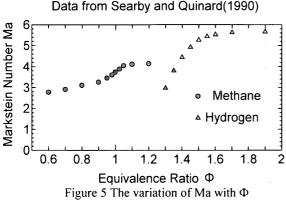


Figure 4 The effects of the dilution on the turbulent velocities



for methane and hydrogen flames[7]

3.4 The mechanisms based on flame stretch

In the following section, the mechanisms of improving the turbulent combustion performance by hydrogen addition will be explained on the basis of the change in the local burning velocity due to the flame stretch. As spherical flame propagates outwardly from the ignition point, the flame area increases with time. Meanwhile each flame element is subjected to the stretch due to the flame curvature and due to the flow field strain rate. In this case, the local burning velocity of each stretched flame element changes from the unstretched laminar burning velocity. Following [6], the general relationship can be expressed as

$$\frac{S_L}{S_{L0}} = 1 - Ma \times Ka \tag{1}$$

Where S_L is stretched laminar burning velocity, S_{L0} is unstretched laminar burning velocity, Ma is Markstein number and Ka is Karlovitz number. Ka is also defined as follows.

$$Ka = \frac{\alpha}{\left(S_{L0}/\delta_L\right)} \tag{2}$$

Where δ_L is laminar flame thickness and α is the flame stretch rate, which is expected to be a positive value on average for outwardly propagating flame. With respect to Ma, there have been various experimental and numerical studies so far [6]-[9], and its general behavior has been clarified to some extent. Generally it has been reported that Ma is mainly related to Lewis number, Le, and changes near-linearly with Le over a wide range of mixtures[6]. Figure 5 shows, as one example, the experimental result on Ma estimated for methane and hydrogen mixtures[7]. From this figure, the methane and hydrogen mixtures have a Markstein number which increases with increasing equivalence ratio.

First of all, how the change in the local burning velocity due to the dependence of Ma on equivalence ratio affects the turbulent combustion performance is discussed for methane mixtures (δ =0) as shown in Fig.2(a). In general, the turbulent burning velocity can be assumed to be product of the flame area and its local burning velocity. Hence assuming that the local flame quenching is less dominant, S_T/S_{L0} can be expressed as

$$\frac{S_T}{S_{L0}} \propto \frac{\bar{S}_L}{S_{L0}} \times \frac{A_T}{A_L} \tag{3}$$

where \bar{S}_{I} is the local burning velocity averaged along the

flame surface and A_T is the total area of laminar surface contained in an area of turbulent flame whose time-averaged area is A_L. Thus A_T/A_L represents the effect of the increase in the flame area by turbulence, and is expected to mainly depend on u'/S_{L0}. Thus from Eq.(3), the difference in S_T/S_{L0} for a constant u'/S_{L0}, as seen in Figure 2(a), is expected to be mainly due to the change in \bar{S}_L/S_{L0} with equivalence ratio. Now the causes of the change in \bar{S}_L/S_{L0} are considered as follows from the viewpoint of flame stretches. For the outwardly propagating spherical flame investigated in this study, each local flame element in the turbulent flame is subjected to a stretch mainly due to;

(A) flame curvatures

(B) strain rate in the flow field.

The latter flow field can be also divided as follows with regard to generative force, that is,

(B-1) unburned gas flow ahead of the flame front induced by combustion

(B-2) turbulent flow field generated by fans.

The flame stretches due to (A) and (B-1) are expected to be mainly depend on the flame topography.

Now in order to investigate the flame stretch due to cause (B-2), Karlovitz number, Ka_{λ} , is introduced, which is defined as

$$Ka_{\lambda} = \frac{\left(\delta_{L}/S_{L0}\right)}{\left(\lambda_{g}/u'\right)} \tag{4}$$

where λ_g is Taylor scale based on the assumption of isotropic turbulence. The relationships between u'/S_{L0} and estimated Ka_{λ} are also shown in Fig.4. From each figure, the differences in Ka_{λ} among the mixtures are appreciable for a certain u'/S_{L0} despite almost the same S_T/S_{L0} . As discussed in the previous paragraph, the mixtures for a specified δ and Φ are expected to have almost the same \overline{S}_L/S_{L0} for a constant u'/S_{L0}. Thus, it follows that the contribution of the flame stretch due to the turbulent flow generated by fans to the change in \bar{S}_L/S_{L0} is relatively small. The main reason is the followings. For the isotropic turbulence, each local flame element in the flow field is expected to suffer both stretch and compression equally. Thus this results in relatively small influences on the average flame stretch, that is, the resultant change in \overline{S}_L/S_{L0} .

From the above discussion, it follows that for the spherical flames propagating outwardly, the flame stretches caused by (A) and (B-1) are primarily responsible for the change in \bar{S}_L/S_{L0} . In this case, the average flame stretch is expected to depend on the flame topography, that is, the distribution of the flame curvature, which is, in turn, expected to depend mainly on u'/S_{L0}. Thus the values of Ka based on the average flame stretch are expected to be almost the same for the mixtures of a constant u'/S_{L0} and intrinsically positive for the spherical flame. Hence in this case, from Eq.(1), the differences in \overline{S}_{L}/S_{L0} mainly depend on the differences in Markstein number among the mixtures. The methane mixtures, as shown in Fig.5, have a Markstein number which increases from about 2.5 in lean mixtures to about 4 in rich mixtures. As a result, it is expected that the methane mixtures have the highest \overline{S}_{1}/S_{10} in leaner region, decreasing with equivalence ratio.

Consequently, the differences of \overline{S}_L/S_{L0} with equivalence ratio due to the dependence of Markstein number on equivalence ratio result in the fact that the leaner mixture has the higher S_T/S_{L0} for a certain u'/S_{L0} , as shown in the Fig.2(a).

Next, as shown in the Fig.3, the mechanism on the change in S_T/S_{L0} with δ for a constant u'/S_{L0} at each equivalence ratio is discussed. From Fig.5, hydrogen mixtures have the highest Markstein number with Ma=6 for very rich regions, decreasing very rapidly for equivalence ratios less than 1.5. This trend is the same as that for methane. Yet in the lean mixture region, the Ma of hydrogen is expected to be smaller than the Ma of methane for a specified equivalence ratio because of smaller Le. For this reason, the hydrogen addition to methane mixture decreases Ma with δ . The decrease in Ma for the lean mixtures causes the relative increase in \overline{S}_L/S_{L0} for a specified u'/S_{L0} and this, in turn, results in a relative increase in S_T/S_{L0} .

On the other hand, in the rich mixture region, Ma of hydrogen is larger than that of methane. Thus for the rich mixtures, the hydrogen addition is expected to increase Ma with δ . As a result, S_T/S_{L0} decreases with δ for a constant u'/S_{L0} because of the relative decrease in \overline{S}_L/S_{L0} . Moreover, because nitrogen is the major component of the mixtures, it is also expected that the Markstein number should principally depend only on the equivalence ratio and not on dilution by nitrogen. Thus it follows that the relationships between u'/S_{L0} and S_T/S_{L0} for the mixtures with the same constituent fuels mainly depend only on the equivalence ratio, not dilution. Thus, as shown in Fig.4, the relationships between u'/S_{L0} and S_T/S_{L0} are almost identical independent of dilution for each δ at a specified Φ .

4. CONCLUSIONS

The following conclusions were obtained in this study. (1) In the lean mixture region, at a certain equivalence ratio, S_T/S_{L0} increases with δ for a constant u^\prime/S_{L0} . The differences decrease as Φ approaches unity. In the rich mixture region, S_T/S_{L0} does not change, or, in some cases, decreases with δ for a constant u^\prime/S_{L0} . Thus considering the balance between improvement of the turbulent combustion performance and the small fluctuation of the operating condition, it can be concluded that the small amount of hydrogen addition is suitable actually for the lean-burn applications.

(2) The relationships between u'/S_{L0} and S_T/S_{L0} for the mixtures with the same constituent fuels are almost identical, independent of dilution at a certain equivalence ratio.

The above effects can be explained by the change in local burning velocity due to the flame stretch as follows;

(3) For a constant u'/S_{L0} , the differences in S_T/S_{L0} with Φ are mainly caused by the changes in the average local burning velocities due to the flame stretch. The changes in the local burning velocity depend on the Markstein number. The methane flames have a Markstein number which increases with equivalence ratio. This trend results in the

highest turbulent burning velocity in the lean mixture region, decreasing with equivalence ratio.

- (4) Because nitrogen is the major component of the mixtures, it is also expected that the Markstein number should principally depend only on the equivalence ratio but not on dilution by nitrogen. Thus the relationships between u'/S_{L0} and S_T/S_{L0} for the mixtures with the same constituent fuels mainly depend on the equivalence ratio, not the dilution.
- (5) In the lean mixture region, the decrease in the Markstein number by hydrogen addition results in the relative increase in \bar{S}_L/S_{L0} for a specified u'/S_{L0}. This, in turn, increases S_T/S_{L0}. In the rich mixture region, on the other hand, the hydrogen addition increases the Markstein number of the methane mixture. This leads to the decrease in S_T/S_{L0} with δ due to the relative decrease in \bar{S}_L/S_{L0} .

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