【Short Paper】

The shape and fluctuation of hydrogen-propane-butane-air lean premixed flames formed on a flat flame burner

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Abstract

In this paper, the characteristics of hydrogen-propane-butane-air lean premixed flames on a flat flame burner were experimentally studied. To elucidate the shape and fluctuation of premixed flames, the cell width, RMS of fluctuation, peak frequency and reconstructed attractor were obtained from the direct photographic images and time series of light emission intensity. The cellular flames were observed at sufficiently low equivalence ratios; the flat flames were found under the other conditions. When the concentration of hydrogen was higher, the flat flames were formed at lower equivalence ratios. As the equivalence ratio became lower, the cell width and RMS increased in the range of cellular flames, which was due to high instability level. At low (high) equivalence ratios, moreover, the size of attractor was large (small) and the trajectory was unstable (quasi periodic). The experimental results revealed the instability characteristics of hydrogen-propane-butane-air lean premixed flames, and the obtained findings were applicable to the safe control of lean premixed flames.

Keywords : Premixed combustion, Hydrogen flame, Liquefied petroleum gas (LPG) addition, Intrinsic instability, Cellular flame, Light emission, Attractor reconstruction

1. Introduction

Nowadays, global warming and air pollution become severe problems, which are mainly caused by emissions of carbon dioxide and nitrogen oxides from the combustion of fossil fuels. Hydrogen is now the focus of attention as an alternative clean fuel. To suppress the emission of environmentally harmful substances, hydrogen lean premixed flames draw more attention as clean energy conversion through combustion. Since hydrogen flames which have high burning velocity and low heating value in volume are colorless, it is difficult to control them safely in energy converters. In addition, the cellular structure and unstable behavior appear owing to intrinsic instability. Thus, we need the visualization of hydrogen flames to control intelligently lean premixed combustion.

To visualize hydrogen flames for safe control, we can add propane and butane which are main components of liquefied petroleum gas (LPG). The addition of LPG to hydrogen causes not only visualization of flames but also low burning velocity and high heating value. Moreover, the combustion of LPG emits low carbon dioxide compared with coal and oil. Thus, the mixture of hydrogen and LPG is one of fabulous candidates for safe and clean fuels.

The mixture of hydrogen and LPG is gathering attention and has investigated by several researchers. The important parameters of premixed flames, i.e. the burning velocity, Markstein length and flame thickness, were experimentally obtained (Miao et al., 2014), and the emission and heat transfer were estimated (Zhen et al., 2012). Moreover, the mixture of hydrogen and hydrocarbon is also gathering attention, and several research papers have been published (Yu et al., 1986; Choudhuri and Gollahalli, 2000; Guo et al., 2005; Halter et al., 2007; Nakahara et al., 2015; Okafor et al., 2016).

The consumption of LPG is rising steadily in Southeast Asian countries, such as Thailand, Vietnam and so on (TechSci Research, 2021). Especially in Thailand, the LPG consumption is expected because of high heating value and low processing cost (EPPO Annual Report, 2019). Thus, the mixture of hydrogen and LPG becomes the fabulous candidate also in Southeast Asian countries. To control safely the combustion of lean mixture of hydrogen and LPG, we need the knowledge on the characteristics of premixed flames. However, the flame instability has not been thoroughly studied, even though the cellular structure and unstable behavior due to intrinsic instability play an important role in the control of premixed combustion. Thus, we shall experimentally study the instability phenomena of hydrogen-LPG premixed flames under the lean conditions by means of the previous method (Kaewpradap and Kadowaki, 2017).

We know three types of intrinsic instability, i.e. hydrodynamic, diffusive-thermal and body-force instabilities (Williams, 1985). In hydrogen lean premixed flames, hydrodynamic and diffusive-thermal instabilities are essentially significant to control the combustion in energy converters. Hydrodynamic instability caused by thermal expansion across the flame front occurs in all premixed flames, and diffusive-thermal instability caused by preferential diffusion of mass versus heat appears in hydrogen lean premixed flames. The experimental and analytical researches on cellular instability of hydrocarbon flames was performed (Sivashinsky, 1983; Konnov and Dyakov, 2007; Yu et al., 2013). Recently, the advanced techniques in laser and computer make it possible to obtain the data elucidating the cellular structure of hydrogen flames. The experiments on laser diagnostics were performed and the essential cellular flame parameters were revealed (Jin et al., 2015; Katsumi et al., 2015), and numerical simulation on the unstable behavior of premixed flames based on the detailed chemical reaction model was performed (Park et al., 2020; Kadowaki et al., 2021).

In this study, mixtures of propane and butane imitating LPG were added to hydrogen with the aim of visualizing hydrogen flames. Hydrogen-propane-butane-air lean premixed flames are formed on a flat flame burner, and the shape and fluctuation were studied. The shape of a cellular flame was identified directly from the photograph, the relationship between the cell width and equivalence ratio was elucidated, and the light emission intensity from hydrogen-LPG premixed flames was analyzed to obtain the RMS of fluctuation, peak frequency and reconstructed attractor for clarifying the characteristics of flame fluctuation. The instability characteristics leading to the safe control of lean combustion of hydrogen were revealed.

2. Experimental procedures

We used the experimental apparatus shown in Fig. 1. Hydrogen-propane-butane-air premixtures through a mixing chamber (MX) were flowed into a flat flame burner with the surface of sintered bronze (McKenna), and premixed flames formed on a porous board with 60 mm in diameter. The burner was water-cooled to adjust the experimental conditions. Pressure reducing valves (RV1, 2, 3 and 4) were installed in each gas tank, and flashback arresters (FA1, 2, 3 and 4) were set. The flow rates of gases were controlled by digital mass flow controllers (MF1, 2, 3 and 4; Azbil, MQV Series).

Fuel gases used in this study were mixtures of propane and butane (imitating LPG) and hydrogen. Considering commercial standard LPG, mixtures of 50% propane and 50% butane were adopted, which were added to hydrogen. The concentration of hydrogen C_{H_2} was varied 98% to 50%. The total flow rate was fixed at 25 L/min. Cellular flames were not observed when the concentration of hydrogen was 50% or less. In addition, we focused on lean combustion, so that the equivalence ratio ϕ was set less than unity.

Images of cellular flames were directly identified by real-time monitoring photography using a digital single-lens reflex camera (Nikon, D5100). The images were taken from the reflection by a mirror placed above premixed flames. The sensitivity of camera was according to ISO 400, the F value was 7.1, and the shutter speed was 1 s.

To measure the light emission from premixed flames, a photodiode (Hamamatsu, S1223-01) was used. The output current from a photodiode was converted to the voltage by a current-voltage conversion amplifier (Hamamatsu, C9329), and the signal was recorded by a data logger (KEYENCE, NR-600, NR-HA08). The sampling frequency and sampling number were 500 Hz and 32678, respectively. The measured signal corresponded to the fluctuation caused by the behavior of entire flames.

The measured emission intensity $x(t)$, varying with time t , was normalized by its average value $\bar{x}(t)$ as follows:

$$y(t) = \frac{x(t) - \bar{x}(t)}{\bar{x}(t)} = \frac{x(t)}{\bar{x}(t)} - 1 \quad (1)$$

Fig. 1 Experimental apparatus

Analyzing the light emission intensity, we used the normalized intensity $y(t)$ to obtain the root mean square RMS, power spectrum density, peak frequency and reconstructed attractor.

3. Results and discussion

3.1 Shape of premixed flames

Table 1 shows the summary of flame shapes, i.e. the range of equivalence ratio ϕ on cellular and flat flames. The cellular flames are observed at sufficiently low equivalence ratios, and the flat flames are found under the other conditions. When the concentration of hydrogen is higher, the flat flames are formed at lower equivalence ratios. Cellular shapes are not observed when the concentration of hydrogen is 50% or less.

Table 1 Summary of flame shapes

C_{H_2}	ϕ (Cellular Flames)	ϕ (Flat Flames)
98	0.25 - 0.40	0.41 -
97	0.27 - 0.41	0.42 -
95	0.30 - 0.43	0.44 -
93	0.33 - 0.45	0.46 -
90	0.37 - 0.47	0.48 -
80	0.44 - 0.49	0.50 -
70	0.50 - 0.53	0.54 -
60	0.52 - 0.54	0.55 -
50	not observed	0.56 -

Fig. 2a Direct photographic images of premixed flames

Fig. 2b Direct photographic images at $C_{H_2} = 90$ and $\phi = 0.44$ (left) and 0.48 (right).

Figure 2a shows the direct photographic images of premixed flames, depending on ϕ and C_{H_2} . Separating cellular and flat flames, the direct photographic images at $C_{H_2} = 90$ and $\phi = 0.44$ and 0.48 are shown in Fig. 2b; a cellular flame is observed at $\phi = 0.44$ and a flat flame at $\phi = 0.48$. Based on these images, the cell width related closely with intrinsic instability is estimated. We count the number of cells along the vertical and horizontal lines (See Fig. 3), and then the cell width W is estimated by the following relation:

$$W = \frac{d}{(N_v + N_h)/2} \quad (2)$$

where d is the diameter of burner surface, N_v and N_h are the number of cells along the vertical and horizontal lines, respectively. This method is simple and versatile to estimate the cell width (Kaewpradap and Kadowaki, 2017).

Figure 4 shows the relationship between cell width and equivalence ratio at $C_{H_2} = 98, 97, 95, 93$ and 90 . The cell width increases as the equivalence ratio becomes lower under the same hydrogen concentration. This is because the diffusive-thermal effects have a destabilizing influence at low equivalence ratios. Under the same equivalence ratio, moreover, the cell width increases as the hydrogen concentration becomes lower, which is qualitatively consistent with the previous experimental results (Jin et al., 2015). It is considered that the instability caused by thermal expansion becomes stronger owing to the decrease of heat loss.

Fig. 3 Estimation of cell width, based on the image of a premixed flame

Fig. 4 Relationship between cell width and equivalence ratio

3.2 Fluctuation of light emission

To study the fluctuation of light emission which is strongly affected by the flame shape, we obtain the time series of normalized emission intensity (Fig. 5) and the power spectrum density (Fig. 6). Peak frequency is found at $f = f_1$ in Fig. 6. This peak frequency corresponds to the typical fluctuation of premixed flames, which is induced mainly by shear flow between the hot burned gas and atmospheric gas. Similar type of fluctuation, caused by Kelvin-Helmholtz instability, is observed in other flames, e.g. Fujisawa et al. (2014).

Figure 7 shows the RMS of fluctuation depending on the equivalence ratio. In the range of flat flames, the RMS hardly changes, on the other hand, the RMS increases as the equivalence ratio becomes lower in the range of cellular flames. This indicates that diffusive-thermal instability plays an important role in the fluctuation of premixed flames. Using the RMS, therefore, we can diagnose the instability of premixed flames qualitatively.

As shown in Fig. 8, the peak frequency f_1 depends on the equivalence ratio and hydrogen concentration. When the flat flames are found, the peak frequency becomes slightly lower as the equivalence ratio decreases. When the cellular flames are observed, the peak frequency becomes lower with a decrease in the equivalence ratio. The results indicate that the appearance of cellular shapes causes the lowness of the peak frequency. We find that the peak frequency drops rapidly during the transition from flat flames to cellular flames.

Fig. 5 Time series of normalized emission intensity at $C_{H_2} = 98$ and $\phi = 0.25$

Fig. 6 Power spectral density of normalized emission intensity at $C_{H_2} = 98$ and $\phi = 0.25$

Fig. 7 RMS of fluctuation depending on the equivalence ratio

Fig. 8 Peak frequency depending on the equivalence ratio

3.3 Attractor reconstruction

To study the characteristics of fluctuation, multi-dimensional attractor is reconstructed by Takens embedding theorem (Takens, 1981). We produce the n -dimensional vector $V(t)$ in the time-delay coordinate,

$$V(t) = [y(t), y(t + \tau), y(t + 2\tau), \dots, y(t + (n - 1)\tau)] \quad (3)$$

The normalized intensity $y(t)$ is defined in Eq. (1). The embedding dimension n is set to 3, and the time delay τ is assumed to be $1/4f_i$. Figure 9 shows the time-delay coordinate system for the attractor reconstruction. The present method of attractor reconstruction from the time series of emission intensity is similar to our previous analysis (Kaewpradap and Kadowaki, 2017).

Figure 10 shows the reconstructed attractors depending on the equivalence ratio at $C_{H_2} = 98, 95, 90$ and 80 . When $\phi = 0.37-0.5$, the reconstructed attractors have a doughnut shape, and this indicates quasi-periodic behavior of fluctuation. In the range of flat flames (See Table 1), the trajectory is not so unstable. As the equivalence ratio becomes lower, the size of attractor increases, corresponding to the increase of RMS. In addition, the trajectory becomes more unstable at sufficiently low equivalence ratios, rising the degree of complexity. When $C_{H_2} = 98$ and $\phi = 0.25$, the attractor demonstrates a chaotic behavior (See Fig. 9 (c), also), which is due to high instability level due to the diffusive-thermal

effects. It was reported that the diffusive-thermal effects at low Lewis numbers cause the chaotic self-motion of cellular-flame fronts (Sivashinsky 1983), and then we believe that the chaotic behavior is caused mainly by diffusive-thermal instability. It is considered that the effects of shear flow on the chaotic behavior are restrictive, because the quasi-periodic behavior is observed at high equivalence ratios. Therefore, the attractor reconstruction is applicable method for understanding the characteristics of flame fluctuation. In addition, it is significant to clarify the relation between the reconstructed attractor and experimental conditions. Clarifying this relation leads to the effective utilization of reconstructed attractor.

Fig. 9 Time-delay coordinate system; (a) Schematic time-delay from normalized emission intensity, (b) Diagram of creating attractor in 3-dimensional field, (c) Reconstructed attractor at $C_{H_2} = 98$ and $\phi = 0.25$

Fig.10 Reconstructed attractors depending on the equivalence ratio at $C_{H_2} = 98, 95, 90$ and 80

4. Conclusion

We have studied the shape and fluctuation of hydrogen-propane-butane-air lean premixed flames formed on a flat

flame burner to obtain the cell width, RMS of fluctuation, peak frequency and reconstructed attractor. The results are summarized as follows:

- The cellular flames are observed at sufficiently low equivalence ratios. The cell width increases as the equivalence ratio becomes lower, which is due to diffusive-thermal instability.
- In the range of flat flames, the RMS of fluctuation hardly changes; in the range of cellular flames, the RMS increases as the equivalence ratio becomes lower. When cellular flames are observed, the peak frequency becomes lower with a decrease in the equivalence ratio. The appearance of cellular shapes causes the lowness of the peak frequency.
- The reconstructed attractors have a doughnut shape, indicating quasi-periodic behavior of fluctuation. The trajectory becomes more unstable at sufficiently low equivalence ratios, rising the degree of complexity.

As a result, we can find the instability characteristics of premixed flames by the flame shape and time series analysis on light emission. It is suggested that the present finding is applicable to diagnose the instability of premixed flames, especially the cellular flames.

In the near future, we will perform the laser diagnostics on hydrogen-LPG premixed flames, taking account of the obtained results mentioned above, and reveal the essence of unstable behavior of cellular shapes induced by intrinsic instability. Moreover, we will change the total flow rate to study the effects of heat transfer from premixed flames to sintered bronze, and clarify the relation between the reconstructed attractor and cellular-flame formation.

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