Dynamic Behavior of Shperically Expanding Flame of H_2/Air/CO_2 Mixture in a Closed Chamber

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1 Introduction

Hydrogen is well known as an alternative fuel of fossil fuels because of no emissions of carbon dioxide and particulates. Meanwhile, hydrogen tends to cause explosions, which include both detonation and deflagration, owing to the wide flammability range and high burning velocity. In an environment and a facility where hydrogen exists, risk assessment is indispensable to reduce and prevent unacceptable risks.[1] In a risk assessment, the flame propagation velocity is one of the most important factors and need to be estimated appropriately. In the conventional method, a propagation velocity of a spherically expanding flame is calculated as the product of laminar burning velocity and thermal expansion ratio, which is derived from the density ratio of unburnt gas to burnt gas. However, lean premixed flames of hydrogen form cellular flame front owing to the intrinsic instability and its propagation velocity is accelerated because of the increase of the flame front area. In a huge space, cellular flame front develops as flame radius increases, and the flame propagation velocity continues to be accelerated. Therefore, a brand-new method considering flame acceleration is necessary to predict actual flame propagation behavior.

Risks of hydrogen present also in a nuclear power plant. Especially, it is well known that hydrogen explosion occurred at the Fukushima Dai-Ichi Nuclear Power Plant in Japan in 2011. In a nuclear power plant, it is important to understand the flow phenomena and flame propagation phenomena of hydrogen because hydrogen is generated from water by radiation degradation and reaction with Zr which is the material of a nuclear fuel cladding. Moreover, in a severe accident, it is known that inside gas of a reactor building contains H_2O vapor, CO and CO_2, which are generated by evaporation of cooling water and by Molten Core Concrete Interactions (MCCI). Therefore, it is necessary that combustion characteristics of multi-component mixture gas are measured and summarized systematically.

Spherically expanding flames of hydrogen have been investigated by several researchers. Kwon et al. performed experiments using C_3H_8/air and $H_2/O_2/N_2$ mixtures varying mixture ratio and initial pressure in a closed chamber, and showed influences of composition and initial pressure on flame acceleration.[2] Wu et al. conducted the experiments of hydrogen flame under pressurized conditions in order to enhance the hydrodynamic instability, and discussed the instability and the flame acceleration characteristics based on

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self-acceleration exponents.[3] Okafor et al. examined the combustion characteristics, such as laminar burning velocity, flame stretch, and flame instability, of H_2/CH_4/air mixture in experiments which was conducted in a closed chamber varying hydrogen concentration and equivalence ratio.[4] Cheikhravat et al. evaluated the influence of H_2O steam on flame propagation characteristics of H_2/air mixture in experiments using a closed chamber.[5] Kim et al. clarified flame propagation characteristics in a large space by carrying out open-air experiments.[6] These experimental studies elucidated a lot of fundamental and important phenomena. However, flame acceleration phenomena in the range of large flame diameter had not been observed because of the small window on the closed chambers, and detailed structure of the cellular flame front had not been measured by open-air experiments because of low spatial resolution. Moreover, the dynamic behavior of spherically expanding flame of hydrogen-based multicomponent mixture has not been understood enough to predict the flame propagation behavior in a large space, such as hydrogen production plant, hydrogen station, and nuclear power plant.

Previously, we established an experimental apparatus including a closed chamber, which has large windows 300 mm in diameter, and observed flame propagation behaviors of H_2/air mixtures in different equivalence ratios by using Schlieren photography. Based on these results, we examined the flame acceleration characteristics and proposed a simple calculation model of the flame propagation velocity.[7] In this study, we added CO2 into the H2/air mixture and conducted combustion experiments of H_2/air/CO_2 mixtures in the closed chamber which has large windows as the first step to understand the flame propagation characteristics of hydrogen-based multicomponent mixture.

2 Experimental

In experiments, we used same closed chamber as our previous study. The closed chamber has four quartz windows, 300 mm in diameter, on the side walls. The internal shape is an intersection of three cylinders and inner volume is 73 litters. To ignite mixture gas at the center of the chamber, two electrodes are inserted into the chamber from the top and bottom. The overall schematic view of the experimental apparatus is illustrated in Fig.1.

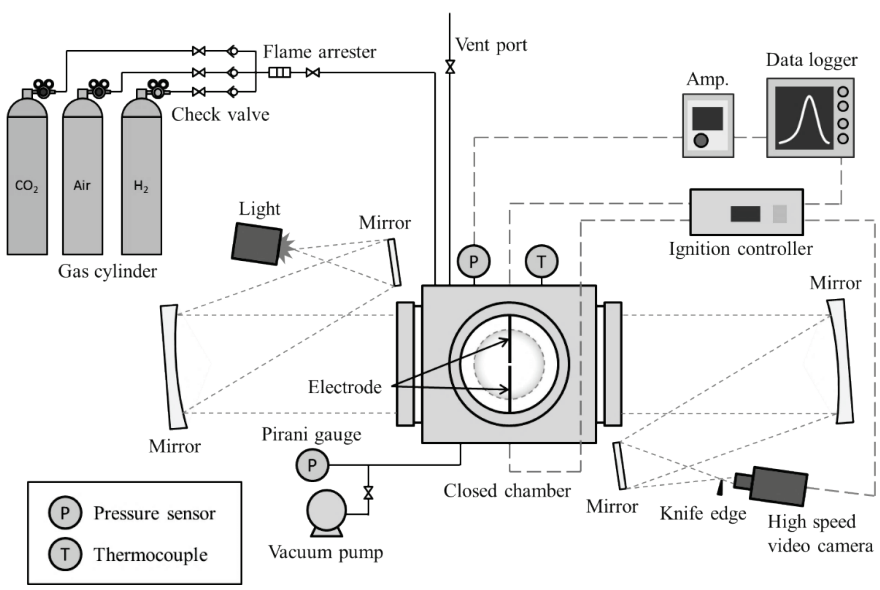


Figure 1. Schematic view of experimental apparatus

The flame propagation behavior was observed by Schlieren photography (Mizojiri SL-350) through the quartz windows and recorded by using a high-speed video camera (Photoron SA-X) which was operated at 10,000 fps with 1024×1024 pixels image resolution. We measured chamber pressure with a capacitive pressure sensor (Kistler 6045A31), and initial temperature with a thermocouple (T-type).

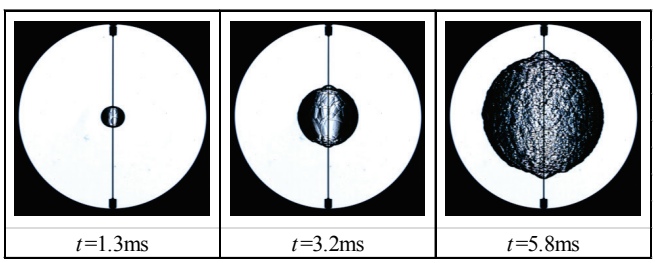
In the preparation of experiments, after vacuuming the chamber, the chamber was filled with air, CO_2, and H_2 up to 101.3 kPa (±0.1 kPa), considering each partial pressure. After that, H_2/air/CO_2 mixture was ignited at the center of the chamber. By a trigger from the ignition controller, the high-speed video camera and the data logger for pressure were synchronized.

In order to evaluate the influence of the mixture composition on the flame propagation characteristics, both of equivalence ratio, φ, and the CO_2 concentration was varied in the range from φ=0.2 to φ=1.0 and in the range from 0 vol.% to 50 vol.%. The Lewis numbers are less than unity in these compositions. At the time of the ignition, Initial pressure, P_0, and initial temperature, T_0, of the mixtures were kept at constant, 101.3 kPa and 298 K, respectively.

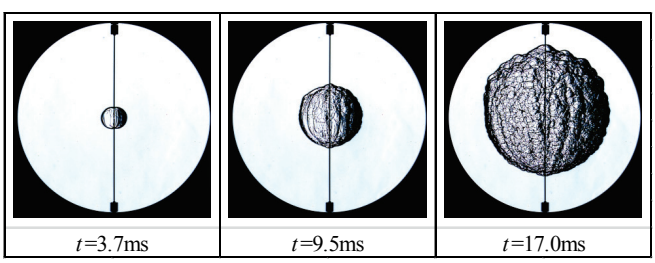
Furthermore, we analyzed images of the obtained Schlieren video frame-by-frame. First, a mean flame radius, r_b, was calculated from area occupied by a flame shadow in a Schlieren image. Next, flame propagation velocity, S_b, was derived by differentiating the flame radius with respect to time. Based upon these values, correlation between flame radius and flame acceleration was obtained finally.

3 Results

Schlieren images of the spherically expanding flames taken by a high-speed video camera are shown in Fig.2. In the both cases shown in Fig.1, cellular structure forms on the flame front due to intrinsic instability, and develops with increasing the flame radius. In the case of mixture including CO_2, cellular structure tends to develop more. This is because the diffusive-thermal effect which is one of factors of intrinsic instability becomes stronger owing to the decrease of burning velocity by CO_2 addition.



(a) CO_2 0 vol.% (r_b=2.0, 5.0, 10 cm)



(b) CO_2 20 vol.% (r_b=2.0, 5.0, 10 cm)

Figure 2. Schlieren images of the expanding spherical flame at φ=1.0

Figure 3 shows the variations of the flame propagation velocity, S_b, with the flame radius, r_b, at the equivalence ratios of 0.6, 0.8, and 1.0. The flame propagation in each case accelerates with increasing the flame radius. This is because the flame surface area increases owing to the development of the cellular flame structure with increasing the flame radius. Moreover, the flame propagation velocity becomes lower at lower equivalence ratio and at higher CO_2 concentration. This is because the laminar burning velocity decreases with decreasing the equivalence ratio and with increasing the CO_2 concentration.

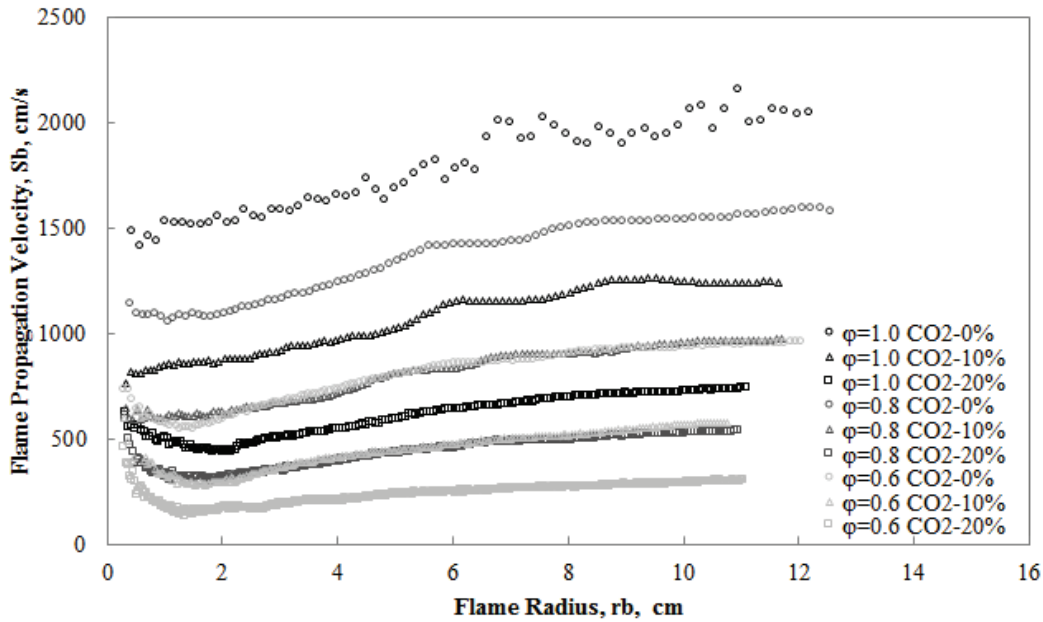


Figure 3. Variations of the flame propagation velocity with the flame radius

4 Discussion

In the range of small flame radius, the flame stretch effect is strong owing to large curvature. The curve expressed by Eq.1, which includes laminar burning velocity, thermal expansion ratio, and a term of flame stretch effect, can fit well the correlation between flame propagation velocity and flame radius in this range. Here, β is propagation velocity of unstretched flame, ρ_u is unburnt gas density, ρ_b is burnt gas density, L is Markstein length, and κ is $2S_b/r_b$. The flame radius where the flame propagation velocity separates from this curve is defined as a critical flame radius, r_0. In the range of larger radius than the critical radius, we used Eq.2 for the curve fitting. Equation 2 includes a term of flame acceleration in addition to Eq.1. We propose Eq.1 and 2 as a brand-new acceleration model.

$$S_b = -\frac{L\kappa}{\rho_u/\rho_b} + \beta \tag{1}$$

$$S_b = \alpha \ln\left(\frac{r_b}{r_0}\right) - \frac{L\kappa}{\rho_u/\rho_b} + \beta \tag{2}$$

By Eq.1, we obtained the propagation velocities of unstretched flame in each case, and these are plotted as a function of equivalence ratio in Fig.4. The propagation velocity of unstretched flame increases with increasing the equivalence ratio, and the propagation velocity of H_2/air mixture shows good agreement with

the data of other researchers. The propagation velocity of H_2/CO_2/air mixture decreases with increasing the CO_2 concentration. This should be because the radiation heat loss increases by CO_2 addition[8].

The critical flame radius, r_0, is summarized as a function of equivalence ratio in Fig.5. In the range of higher equivalence ratio than around 0.5, the critical flame radius decreases with decreasing the equivalence ratio and with increasing the CO_2 concentration. This could be because the diffusive-thermal effect becomes strong owing to the decreases of the effective Lewis number and the burning velocity.

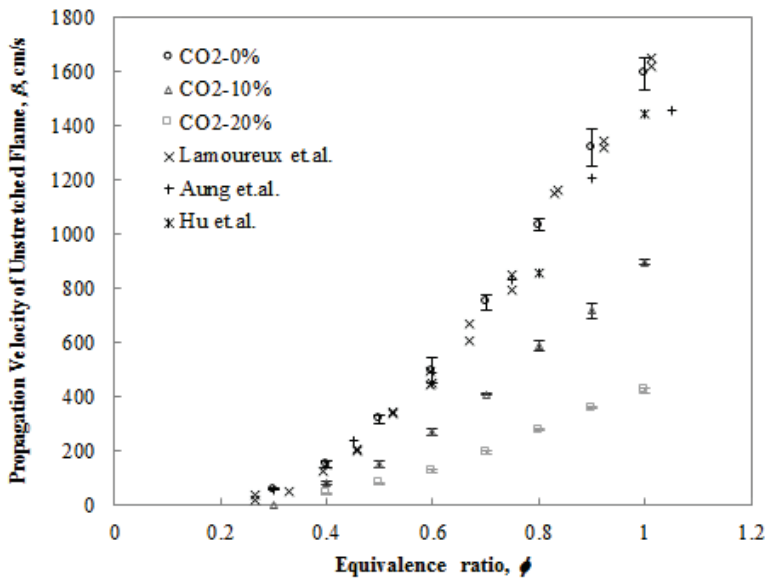


Figure 4. Correlation between flame propagation velocity of unstretched flame and equivalence ratio[9-11]

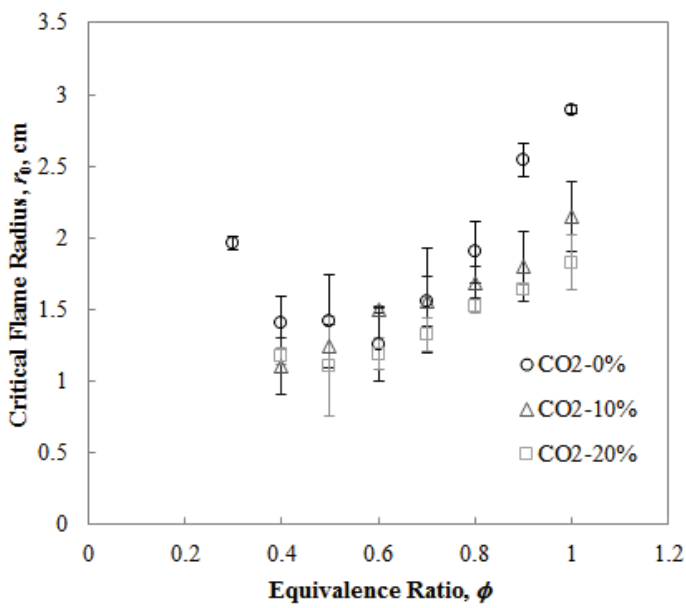


Figure 5. Correlation between critical flame radius and equivalence ratio

5 Conclusion

Spherically expanding flame of H_2/air/CO_2 mixture was observed by Schlieren photography in a closed chamber. The equivalence ratio and CO2 concentration were varied in order to evaluate the influence of the mixture composition on the flame propagation behavior. As the results, we found the following facts: (1) the cellular flame structure develops more at high CO_2 concentration because the diffusive-thermal effect becomes stronger, (2) flame propagation velocity of unstretched flame decreases with decreasing the equivalence ratio and with increasing the CO_2 concentration, (3) the critical flame radius becomes smaller with decreasing the equivalence ratio and with increasing the CO_2 concentration.

Acknowledgements

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