Effects of hydrogen addition on flame propagation and blast wave generation during explosion of methane-air mixtures

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1 Introduction

A large number of accidental gas explosions have happened every year and caused indeed serious damage. The potential risk of the explosions has existed in all facilities using combustible gases, such as in chemical plants, high pressure gas storage facilities, and every household using gas fuels. To minimize the risk of accidental gas explosions, the consequence analysis of the possible damages by an accidental explosion has to be performed. The main damages by accidental explosions are often caused by pressure increase, blast wave, fragment scattering and hot gas. The damage caused by a blast wave (pressure wave) can spread quickly and widely, and it could be a significant consequence of the accidental explosion around the explosion point [1]. The understanding of the blast wave intensity from gas explosions is essential to perform the consequence analysis appropriately. In this study, experiments were performed in an open space with the combustible mixtures of various burning velocity and Lewis number, adding hydrogen in methane/air mixtures. The effects of hydrogen addition on flame propagation behavior and blast wave intensity were examined.

2 Experiments

To measure flame propagation behaviors and the blast wave intensity simultaneously in an open space, we prepared the experimental apparatus comprising the ignition system, the high speed schlieren photography system and the acoustic measurement system. A spherical soap bubble with hydrogen /methane/air mixtures was made by premixed combustible gas flow nozzle. When a gas explosion was triggered in the soap bubble system, the flame propagation sequence in an open space was imaged with schlieren photography and recorded using a high speed camera. The blast wave in an open space was simultaneously measured by microphone, which is 0.3, 0.1m away from the ignition point. Experiments were conducted with hydrogen addition in methane/air mixtures. The effective fuel/air equivalence ratio is defined as

$$\phi_F = \frac{C_F / [C_A - C_H / (C_H / C_A)_{st}]}{(C_F / C_A)_{st}}$$

where $(C_F/C_A)_{st}$ is the stoichiometric fuel-to-air molar ratio, $(C_H/C_A)_{st}$ is the stoichiometric hydrogento-air molar ratio, ratio, $C_F C_H C_A$ are mole fractions of the fuel, hydrogen, and air are, respectively. Hydrogen faction(α_H) is defined as[2]

$$\alpha_{H} = \frac{C_{H} + C_{H} / (C_{H} / C_{A})_{st}}{C_{F} + [C_{A} - C_{H} / (C_{H} / C_{A})_{st}]}$$

3 Results and discussion

Figure1 shows the schlieren photographs of methane/hydrogen/air mixtures at various hydrogen fractions($\phi = 0.7, 1.0, 1.3$). The results show that the flames at equivalence ratios of 0.7, 1.0, 1.3 were intensively wrinkled and accelerated, with hydrogen addition increases in methane/air mixtures. In the present experiment, methane/hydrogen/air mixtures were quiescent at the time of ignition and there was no obstacle in the space where the flame propagated. Therefore, the flame wrinkling might be caused by some kind of flame front instability. One possible generating mechanism of the flame wrinkling is diffusive thermal instability [3]. This instability is driven by differences between the diffusivities of mass and heat and is quantified by the Lewis number. Since there are two fuels in the mixture, the Lewis number should be a weighted average value. In this study, as proposed by Law et al, the Lewis number of fuel is evaluated from the following:

$$Le_F = 1 + \left[\frac{q_1(Le_1 - 1)q_2(Le_2 - 1)}{q_t}\right]$$

where Le_1 , Le_2 are the Lewis numbers of hydrogen/air mixture, Lewis numbers of methane/air mixture, respectively, the nondimensional heat release associated with the consumption of species *i*, defiend as $q_i = QY_i/C_p T_u$, where *Q* is the heat of reaction, Y_i is the supply mass fraction of species *i*, C_p is the specific heat of the unburned gas and T_u is the unburned gas temperature [4]. The effective Lewis number is defined as the combination of the excess and deficient Lewis number, defined as

$$Le_{eff} = 1 + \frac{(Le_E - 1) + (Le_D - 1)A}{1 + A}$$

where Le_E and Le_D are the Lewis number of excessive and deficient reactants, respectively. Here, the parameter $A=1+\beta(\Phi-1)$ is a measure of themixture's strength, which Φ is defined as the ratio of the mass of excess-to-deficient reactants in the fresh mixture relative to their stoichiometric ratio and $\beta = E_a (T_a - T_u)/R^0 T_a^2$ is the Zeldovich number, with T_a the adiabatic flame temperature, T_u the temperature of the fresh mixture, $E_a = -2R^0 [\partial \ln(\rho_u S_u^0)/\partial(1/T_a)]$ the activation energy, and R^0 the gas constant [5].



Figure 1. Schlieren pictures of methane/hydrogen/air mixtures at various hydrogen fractions. ($\phi = 0.7, 1.0, 1.3, t = \text{time from ignition}$)



Figure 2. Left: The effective Lewis numbers and the Lewis numbers of fuels versus hydrogen fractions. Right: measured peak positive pressure versus hydrogen fractions at ϕ of 0.7, 1.0, 1.3.

Figure 2 (left) shows the calculated the effective Lewis number and the Lewis numbers of fuels of versus hydrogen fractions at equivalence ratios of 0.7, 1.0, 1.3. Although the Lewis numbers of fuels of equivalence ratios of 1.3 increase with hydrogen fractions, the effective Lewis number at the overall equivalence ratio increasingly decreased with hydrogen fractions, since the effective Lewis number includes burning velocity and the adiabatic flame temperature. The disposition which the effective Lewis number decreased with hydrogen addition indicates that the flame is cellulary unstable with hydrogen fractions. Figure 2 (right) shows measured peak positive pressure versus hydrogen fractions at equivalence ratios of 0.7, 1.0, 1.3. The overpressure at various equivalences also increase with hydrogen fractions. These results could understand by comparison with exiting model by acoustic theory. The blast wave history p(t) at a distance, d, from the ignition position can be expressed by the acoustic theory as the following equation:

$$p(t) = \frac{\rho}{d} (\varepsilon - 1) \left\{ 2r\varepsilon S^2 + r^2 \frac{dS}{dt} \right\}$$

where ρ is the density of the medium, r is the flame radius, ε is volumetric expansion ratio, S is burning velocity [6]. This model shows that the blast wave depends on the volumetric expansion ratio and square of the burning velocity the flame acceleration. The second term in the curly brace in the right hand side of the equation indicates the contribution of the acceleration of burning velocity. Figure 3 shows that measured overpressure compared with the values calculated from equation by the acoustic theory and time histories of burning velocity at different effective Lewis number. The burning velocity of existing literature [7] was used to calculate the overpressure at a constant burning velocity while the result of the present study was used to calculate the overpressure at acceleration of burning velocity. In a $Le_{eff} < 1$ flame (left), burning velocity accelerated by diffusive-thermal instability, since the combustion reaction is intensified at the convex segment and weakened at the concave segment, leading to smoothing of the wrinkles. Since the overpressure in a $Le_{eff} < 1$ flame (left) increased with the acceleration of burning velocity by diffusive-thermal instability, the measured overpressure is much larger than the evaluated value at constant burning velocity. Consequently in a $Le_{eff}>1$ flame (right), burning velocity is constant without wrinkled flame up to 0.038s, before the burning velocity in the later stage is acceleratingly increased. The measured overpressure in a $Le_{eff} > 1$ flame (right) agreed with evaluated value at constant burning velocity right before appearance of flame instability and acceleratingly increased in the later stage. The flame wrinkling in later stage of rich mixture is generated when the flame approaches to the boundary between the mixture and surrounding air. Therefore, the rupture of soap bubble and heterogeneous mixing of the mixture with the surrounding air will be concerned. That is, the flame wrinkling in rich mixture might be generated by nonuniformity of concentration distribution of combustible gas. In this case, not only the burning velocity



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Figure 3. Measured overpressure compared with the values calculated from equation by the acoustic theory and time histories of measured burning velocity. Left : $\phi = 0.7$, $\alpha_H = 0.6$, $Le_{eff} = 0.608$. Right : $\phi = 1.3$, $\alpha_H = 0.4$, $Le_{eff} = 1.121$. $S_l =$ burning velocity of existing literature, $S_m =$ measured burning velocity.

became a larger value, but also the burning velocity is acceleratingly increased. From this experiment, it is understood that the intensity of the blast wave is strongly affected by both the acceleration of the burning velocity from diffusive-thermal instability and the flame acceleration from during the mixing process of rich combustible mixture with air.

4 Conclusions

To understand the consequence of accidental gas explosions in an open space, the effects of flame propagation behavior on the blast wave were examined experimentally. The spherically propagating flames in the mixtures of methane and hydrogen with air were focused. The results show that the flames at equivalence ratios of 0.7, 1.0, and 1.3 were intensively wrinkled and accelerated with the increases of hydrogen addition in methane/air mixtures. The overpressure of the blast wave tends to increase acceleratingly by the flame wrinkling. The flame winkling is considered to be generated by diffusive-thermal instability according to the analysis of the effective Lewis number. The overpressure can be predicted by the acoustic theory which indicates that the intensity of blast wave is affected, in particular, by burning velocity, volumetric expansion ratio and the flame acceleration. Especially, the intensity of the blast wave is strongly affected by the acceleration of the burning velocity.

References

[1] Dobashi R, Kawamura S, Kuwana K, Nakayama Y. (2011) Consequence analysis of blast wave from accidental gas explosions. Proc. Combust. Inst. 33: 2295.

[2] Yu G, Law CK, Wu CK. (1986) Laminar flame speeds of hydrocarbon + air mixtures with hydrogen addition Combust Flame. 63: 339.

[3] Williams FA. (1985) Combustion Theory 2nd Edition, Westview Press.

[4] Law CK, Jomaas G, Bechtold JK. (2005) Cellular instabilities of expanding hydrogen/propane spherical flames at elevated pressures: theory and experiment. proc. Combust. Inst. 30: 159.

[5] Addabbo R, Bechtold JK, Matalon M. (2002) Wrinkling of spherically expanding flames. proc. Combust. Inst. 29: 1527.

[6] Thomas A, Williams GT. (1966) Flame noise : Sound emission from spark-ignited bubbles of combustible gas, Proc. Roy. Soc. A, 294: 449.

[7] Hu E, Huang Z, He J, Jin C, Zheng J. (2009) Experimental and numerical study on laminar burning characteristics of premixed methane–hydrogen–air flames Int. J. hydrogen eng. 34: 4876.