Experimental study on the explosion characteristics of methane-hydrogen/air mixtures

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1 Introduction

Natural gas (NG) is being widely recognized as one of the most promising alternative fuels due to its large reserves, low CO_2 and pollutants emission in combustion and high octane number in engines [1]. For now, in many situations, NG has already been used and promoted in industrial production, municipal engineering, internal combustion engine, and household appliance.

In the meanwhile, hydrogen-enrichment is regarded as a useful tool to overcome some drawbacks in NG combustion system [2], such as local flame extinction, combustion instability, lower power output, etc.. Besides, it is proved that, hydrogen addition can significantly enhance the combustion and flame dynamics through both thermal and chemical effects [3]. Nevertheless, the defect of hydrogen-NG system is obvious. That is, natural gas consists mainly of methane, which is highly flammable and explosive. Then due to the unique properties of hydrogen, such as wider flammability ranges, higher diffusivity, lower ignition energy and larger burning velocity, it would probably increase the sensitivity and severity of explosion of gas mixtures. Therefore, as the development bottleneck of hydrogen-NG system, the concomitant safety issues must be fully evaluated for helping make laws and regulations, and improving the explosion prevention and protection designs.

In general, the explosion limits, maximum explosion pressure, maximum rate of pressure rise and combustion duration are definitely the most important parameters to characterize and quantify the explosion risk and consequence [4, 5]. Previously, most of the attention was paid to the explosions and fundamental combustion properties of natural gas (or methane). Only few of the literature talked about the hazards of hydrogen-natural gas (methane) mixtures. Cammarota et al. [6]experimentally studied and compared the explosion behaviors of stoichiometric hydrogen-enriched methane/air (with 10% Hydrogen in the fuel) and methane/air mixtures. Then stoichiometric (Φ = 1) hydrogen-methane mixtures with varied hydrogen content at three initial pressures (1, 3, 6 bar) were examined to reveal the combined effect of fuel composition and initial pressure on explosion [7]. Ma et al. [8] and their group have also done some pioneering work on basic explosion parameters of hydrogen-methane mixtures with different hydrogen

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concentration at only three equivalence ratios. Besides, they recently investigated and compared the confined and vented explosions of stoichiometric hydrogen-methane mixtures [9]. Other than these small and medium scale explosions, the large scale experiments and simulations have been carried out as well under the European Commission (EC) –funded project NatrualHy to study the vented and confined explosions with/without congested region for assessing the explosion consequences of potential hydrogen-methane mixtures in the pipeline networks [10]. In addition, some previous works concentrated on the premixed hydrogen-methane/air deflagrations and flame propagations [11].

In summary, the explosion data of hydrogen-natural gas (methane) is still very limited. The explosion characteristics at wider equivalence ratio range should be scrutinized further. And deep insight to the rules of all the explosion phenomenon and data is necessary for assessing the hazards of energy system and guiding the fuel preparation and engine safety design. Therefore, in present study, experiments were systematically performed in a standard 20-L explosion spherical vessel for different methane-hydrogen/air mixtures. The pressure dynamics was recorded and analyzed to obtain detailed explosion and combustion parameters.

2 Experimental method

The 20-L explosion spherical vessel with 16.84 cm inner diameter was built according to ISO6184-1 for measuring explosion parameters of gas or dust mixtures (see Fig.1). The experimental system also contains an ignition device, a control unit, a data acquisition system, and a vacuum pump.

The vessel was vacuumed first in the beginning and then filled with gases by partial pressure method until the target composition was reached. The purity is 99.9% for both methane and hydrogen gases. The initial temperature and pressure were 298 K and 101,325 Pa, respectively.

The quiescent mixture was centrally ignited (10 J energy) by the pair of electrodes. And the subsequent explosion process was monitored by a PCB pressure sensor installed on the wall of vessel. The determination of flammability limits is based on the criterion of 7% pressure rise in the vessel.

Cases were repeated a few times to reduce the random error and ensure the reliability, especially when near the flammability limit (LFL and UFL).

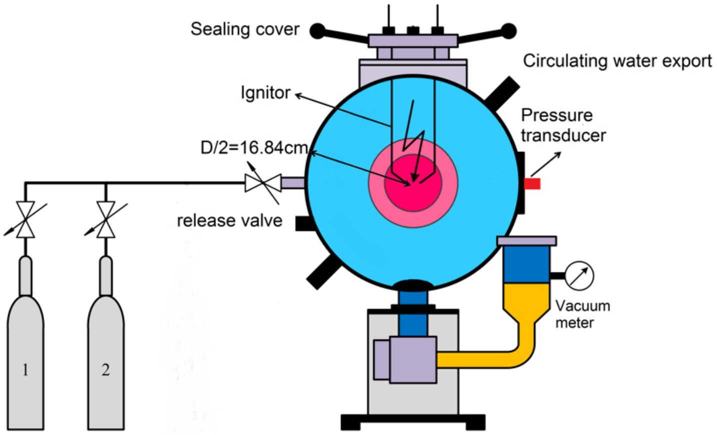

Figure 1. Experimental apparatus

3 Results and discussion

3.1 Maximum explosion pressure and flammability limit

Fig. 2 presents the maximum explosion pressure, P_{max} of different methane-hydrogen/air mixtures. The curves are simulation results by chemical equilibrium method using GASEQ software which is based on the hypothesis of adiabatic expansion. For methane-air mixtures, due to the heat loss and non-equilibrium reaction at off-stoichiometric conditions, the experimental measurements in the vessel are lower than simulation. Nevertheless, near stoichiometric condition or with large contents of hydrogen addition, parts of the experimental data are larger than those from GASEQ. This may be due to some underlying transient effects (chemical or thermal) associated with explosion in the vessel [8].

All the flammability limits of methane in mixtures are summarized in Table 1. Evidently, with hydrogen enrichment, less methane (or even no methane) is needed for a successful explosion. From this point of view, hydrogen addition indeed significantly raises the explosion risk of the binary fuel blend system.

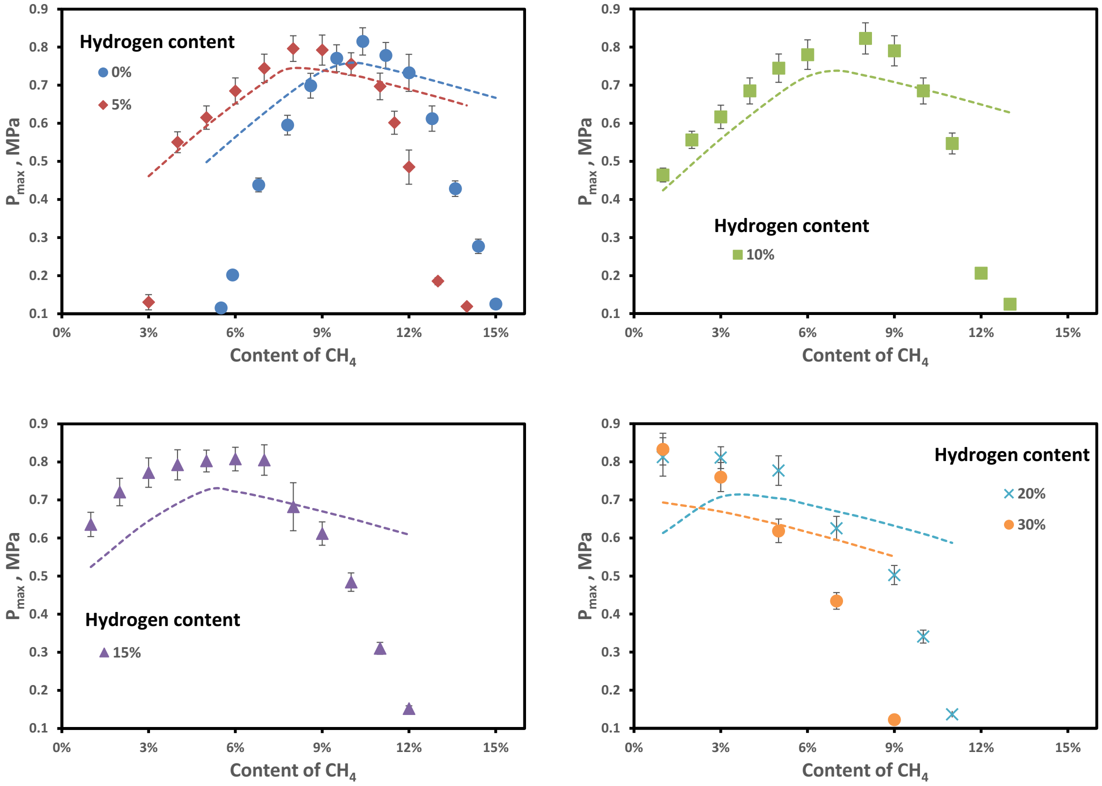

Figure 2. Maximum explosion pressure of different methane-hydrogen/air mixtures

(Symbol: experimental data; Dash line: simulation from GASEQ; The hydrogen content is the same for both symbol and line with the same color)

Table 1: Flammability limits of methane in the mixtures with various hydrogen additions

H_2 (%vol.)	LFL(CH_4) (%vol.)	UFL(CH_4) (%vol.)
0	5	15
5	3	14
10	-	13
15	-	12
20	-	11
30	-	9

1. "-" means the LFL has no meaning in that condition.
2. Based on the previous criterion of a 7% pressure rise, the LFL ≃ 6.0±0.5% for hydrogen/air in the 20-L chamber. [4]

3.2 Maximum rate of pressure rise and deflagration index

Fig. 3 shows the maximum rate of pressure rise, $(dP/dt)_{max}$ of different methane-hydrogen/air mixtures. Unlike P_{max}, the peak value of $(dP/dt)_{max}$ for each curve differs and grows up with hydrogen content. In other word, hydrogen addition can effectively promote the pressure growth rate in the vessel, which is very likely due to the increment of laminar flame speed by hydrogen in the mixture.

The deflagration index, K_G is calculated from $(dP/dt)_{max}$ and vessel volume, which is another important explosion parameter manifesting the intrinsic property of the mixture, and is independent of the explosion apparatus used in the experiments. Therefore, in Fig.4 we compared experimental and simulation data of K_G from different research groups [5, 7, 8]. Herein α is defined as the volumetric ratio of hydrogen over total fuel (hydrogen/methane mixture). From pure methane to pure hydrogen, α varies from 0 to 1. It could be seen that, our data are consistent with other experiments when $\alpha < 0.8$, but lower than simulation due to the heat loss effect. When $\alpha > 0.8$, our data agree well with simulation, which is due to the hydrogen addition effect on the laminar flame speed: with relatively more hydrogen addition in the fuel, the laminar flame speed is relatively higher as well, which would remarkably shorten the combustion duration and decrease the heat loss to the vessel wall during explosion.

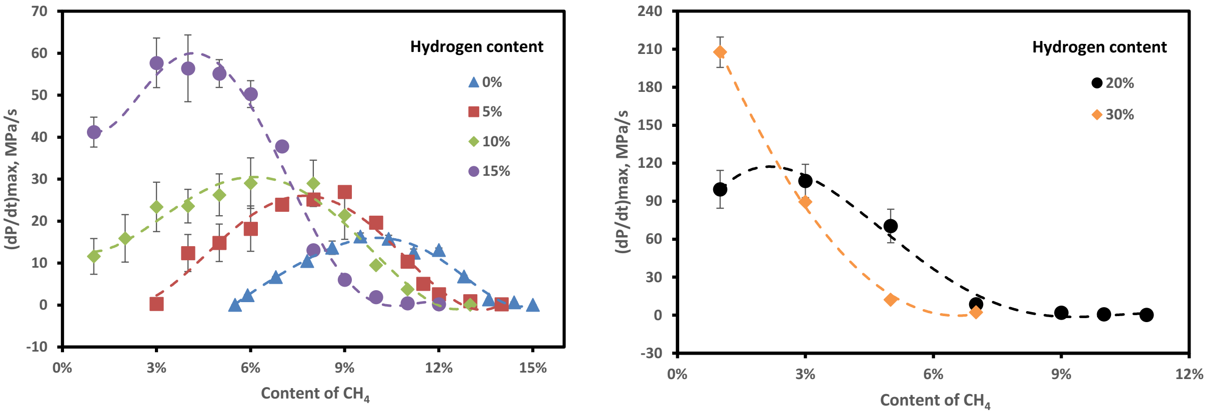

Figure 3. $(dP/dt)_{max}$ of different methane-hydrogen/air mixtures

(symbol: experimental data; dash line: curve fit)

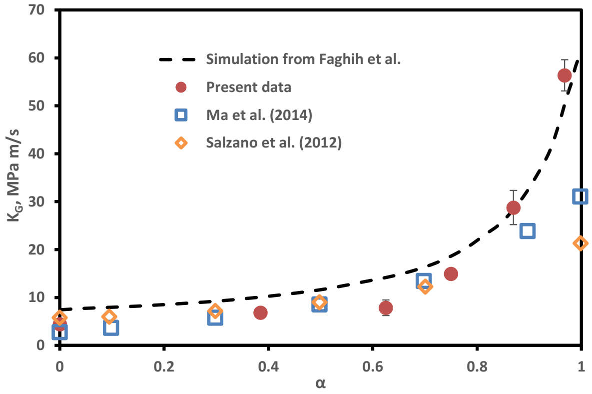

Figure 4. Deflagration index of methane-hydrogen/air mixtures at stoichiometric condition

In general, taking both P_{max} and $(dP/dt)_{max}$ into account, the hydrogen addition can remarkably shorten the combustion time and accelerate the pressure buildup in the confined space. Thus, from the view of safety, for fuel and engine design in industry, energy system with more hydrogen does have higher hazard.

3.3 Laminar flame speed

Compared to previous studies using advanced techniques (counter-flow flame, flat flame, expanding spherical flame method, etc.) [3], in this work the laminar flame speed was derived from the pressure history during explosion in the vessel with a theoretical model proposed by Dahoe [12]. The method was considered as an acceptable conservative estimation for engineering calculation and thus was adopted and validated in this work.

Fig.5 summarizes the laminar flame speeds of various methane-hydrogen/air mixtures measured in this work. Evidently, as hydrogen increases in the mixture, the curve moves to the left with the peak value growing up notably, which is quite similar to $(dP/dt)_{max}$ discussed above. Compared to methane, hydrogen has much smaller molecular weight, larger thermal conductivity, higher reactivity and flame temperature, which explains the acceleration effect on the flame propagation with hydrogen addition in the mixture.

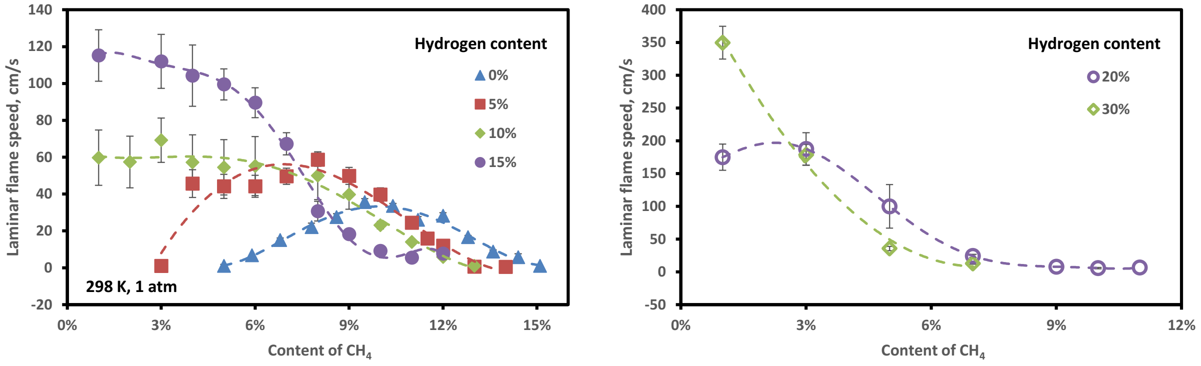

Figure 5. Laminar flame speeds of different methane-hydrogen/air mixtures

4 Conclusions

Explosion and combustion parameters of various methane-hydrogen/air mixtures were revisited and measured in a wider range of concentrations in a standard 20-L spherical vessel with pressure sensor. Some conclusions are drawn as follows:

(1) The hydrogen addition could significantly decrease the LFL and UFL of methane in the mixture, remarkably shorten the combustion time and accelerate the pressure buildup in the confined space.

(2) Unlike P_{max}, the peak values of the maximum pressure rise rate, $(dP/dt)_{max}$ versus methane content grow up prominently with hydrogen content due to the increment of laminar flame speed.

(3) The laminar flame speed of methane-hydrogen/air mixture varies in a similar way to $(dP/dt)_{max}$ and is mainly controlled by the total equivalence ratio Φ_{total} of the methane-hydrogen/air mixture. Dahoe's model is a feasible way to assess the laminar flame speed with low cost but acceptable accuracy.

References

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