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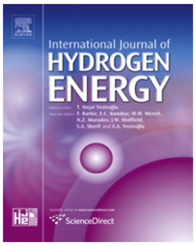

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Flammability limits of hydrogen-enriched natural gas

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ABSTRACT

This paper reports both the lower and upper flammability limits of hydrogen-enriched natural gas with hydrogen fractions of 20%, 40%, 60% and 80% respectively as well as these of natural gas and hydrogen, measured by using a constant volume combustion chamber together with a high-speed schlieren photographic system. Based on investigating pressure rise history inside the combustion chamber as well as flame photos, the effect of hydrogen enrichment on the flammability characteristics is discussed. Our experimental results show that the flammability limits of methane–hydrogen mixtures can be used for hydrogen-enriched natural gas as long as their hydrogen fractions are the same. In this paper, the flammability data of methane–hydrogen mixtures available in the literature are reviewed. Correlations for both the lower and upper flammability limits of methane–hydrogen mixtures are summarized.

1. Introduction

Hydrogen is regarded as "an ideal fuel from the point of conservation of the environment" because "the only toxic products of combustion of hydrogen are nitric oxides" [1]. The recent efforts on solar-hydrogen technology lead its way to turn hydrogen energy into a promising renewable energy resource (e.g. [2–4]). This adds measures for the large-scale usage of hydrogen energy in the future. But with our current technologies, widespread application of pure hydrogen in transport engines is still unlikely to happen in the near future, mainly due to infrastructure, transportation and storage constrains [1].

On the other hand, natural gas has been used widely in transportation and industry as well as in domestic applications. To increase its thermal efficiency and reduce unburned hydrocarbon emissions at lean operation conditions, one promising method is to add hydrogen (whose burning velocity is six to seven times as fast as that of natural gas) into natural gas. Experiments showed that by fueling hydrogen-enriched natural gas, automotive engines can operate smoothly at lean conditions with improved engine performance, increased thermal efficiency and reduced emissions (e.g. [5–7]). Therefore, hydrogen-enriched natural gas provides a feasible solution for the high-efficient and environmentally friendly usage of both hydrogen and natural gas.

To safely use hydrogen-enriched natural gas, the knowledge on the explosion hazards of these mixed gaseous fuels is of great importance. Flammability limit (also known as explosion limit) has been widely used as an index for the quantitative risk assessment of the explosion hazard associated with the usage of these fuels. There are two flammability limits named as lower flammability limit (LFL) and upper flammability limit (UFL), referring to the leanest and the

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Nomenclature

LFL	lower flammability limit
p	pressure inside the combustion chamber, MPa
p_i	initial pressure, MPa
p_{max}	maximum pressure inside the combustion chamber, MPa
S_N	standard deviation of experimental and calculated flammability limits data
t	time, s
UFL	upper flammability limit
x	volumetric fraction of hydrogen in hydrogen-enriched natural gas
x_1	measured flammability limits of hydrogen-enriched natural gas
x_2	measured flammability limits of methane–hydrogen mixtures
x_3	calculated flammability limits of methane–hydrogen mixtures
$\bar{x}$	mean value of x_1, x_2 and x_3
Δp	pressure rise inside the combustion chamber $\Delta p = p - p_i$

richest fuel/air mixtures upon which a self-sustainable flame can be initiated respectively.

The flammability limits of methane and hydrogen have been measured and reported intensively [8–16]. For methane–hydrogen mixtures, the available flammability data are relatively limited comparing with that of methane or hydrogen. Table 1 summarizes the available flammability limits of methane–hydrogen mixtures reported in Refs. [17–22]. We noticed that the hydrogen fractions of methane–hydrogen mixtures were different in these references. For example, the hydrogen volume fractions studied were 25%, 50% and 75% in Ref. [19], while in [21] the hydrogen fractions were 20%, 40%, and 60% (See Table 1). Wierzba and Ale studied a wide range of fuel mixtures involving hydrogen, covering hydrogen volume fractions of 20%, 50%, 70%, and 90% [18]. However, only the upper flammability limits were reported. Therefore, it is necessary to conduct a systematic experimental study on the flammability limits of methane–hydrogen mixtures.

We also noticed that it is natural gas that has been used widely in transportation and domestic applications, not pure methane. Although methane is the main constitute of natural gas, are the methane–hydrogen flammability data good enough to represent these of hydrogen-enriched natural gas? Is it appropriate to ignore the effects of other constitutes of natural gas on its combustion under hydrogen-enriched environment? Experimental proof is needed to answer these questions. Therefore, we investigated the flammability characteristics of both hydrogen-enriched natural gas and methane–hydrogen mixtures experimentally in this study. Hydrogen-enriched fuels with hydrogen fractions of 0%, 20%, 40%, 60%, 80% and 100% were tested, aiming at providing a complete picture of the effect of hydrogen addition on its flammability limits.

Generally speaking, there exist two types of apparatus for measuring the flammability limits of a fuel. One is stainless steel or glass tube, usually cylindrical with internal diameter of 5–10 cm [9,14–22]. Usually a mixture is treated as non-flammable if its flame fails to propagate a certain length of distance. The other is spherical or cylindrical internal shaped combustion chamber (also referred to as combustion bomb or explosion vessel) with wide range of internal volume from 1.57 dm^3 (also known as liter) up to 25.5 m^3 [10–13,15,16,19,20]. Spark ignition systems or pyrotechnic igniters are needed to ignite the combustible mixtures; ignition energy should be chosen with care, depending on both chamber dimension and properties of the combustible mixtures. Flammability limits can be determined either by pressure rise criterion or visual criterion. The latter requires one or more glass windows in the combustion chamber.

To standardize test method as well as calculation procedure for measuring the flammability limits of gases and their mixtures, both international and national standards are available. A comprehensive review of international standard,

Table 1 – Flammability limits of methane–hydrogen mixtures determined by various methods [17–22].

2.7 L chamber[b]				
Hydrogen fraction (%)	0	25	50	75
LFL (vol.%)	4.6[a]	4.5[a]	4.4[a]	4.2[a]
UFL (vol.%)	16.5[a]	23[a]	32[a]	43.2[a]

4.2 L sphere[c]				
Hydrogen fraction (%)	0	20	40	60
LFL (vol.%)	4.6	4.4	4.6	4.6
UFL (vol.%)	16.0	19.6	25.4	–

Tube with diameter of 50 mm[d]		
Hydrogen fraction (%)	23.08	50
LFL (vol.%)	5.0	4.63

Tube with diameters in the range of 18.4–50.2 mm[e]					
Hydrogen fraction (%)	0	10	20	30	40
LFL (vol.%)	5.1	5.21	4.54	4.48	4.35

Tube with diameter of 50.8 mm[f]						
Hydrogen fraction (%)	0	20	50	70	90	100
UFL (vol.%)	15[a]	18[a]	26[a]	35[a]	51[a]	76[a]

Tube with diameter of 60 mm[g]				
Hydrogen fraction (%)	0	20	40	60
LFL (vol.%)	4.4	4.2	4.0	4.0
UFL (vol.%)	15.8	19.0	24.2	32.4

a Values were read from figure(s).
b Pahl (1994), using 10% pressure rise criterion, from Ref. [21].
c Van den Schoor et al (2008), using 5% pressure rise criterion [21].
d Flammability tube (stainless steel) with diameter of 50 mm, vertical upward flame propagation using visual criterion [17].
e Flammability tubes (pyrex) with diameters in the range of 18.4–50.2 mm, vertical upward flame propagation using visual criterion [22].
f Flammability tube (stainless steel) with diameter of 50.8 mm, vertical upward flame propagation using visual criterion [18].
g Flammability tube (glass) with diameter of 60 mm, vertical upward flame propagation using visual criterion [19–21].

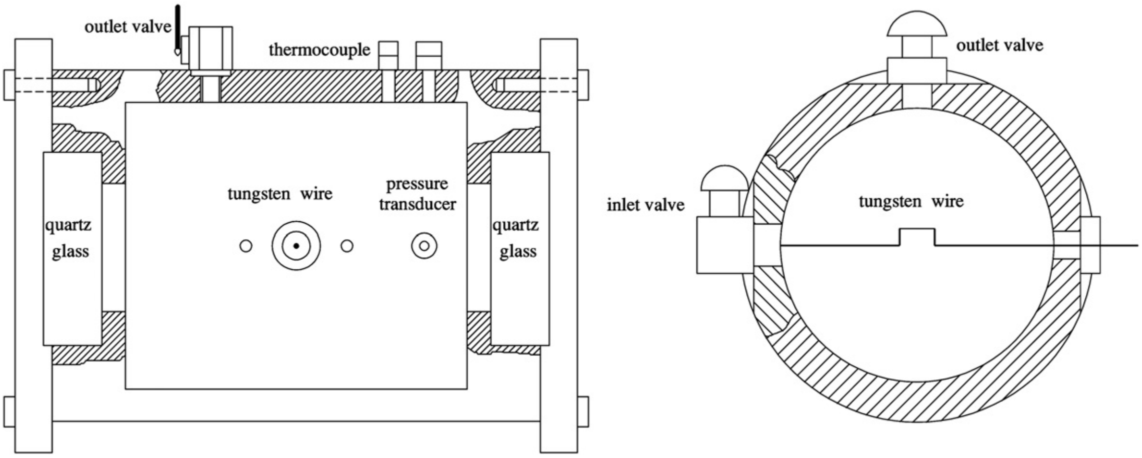

Fig. 1 – The structure of the constant volume combustion chamber.

Table 2 – Constitution of natural gas.

Items	CH_4	C_2H_6	C_3H_8	N_2	CO_2	Others
Volumetric fraction (%)	96.160	1.096	0.136	0.001	2.540	0.067

European standard EN 1839, German standard DIN 51649-1 and US standard ASTM E 681-01 can be found in Ref. [16]. In this study, a combustion chamber with two quartz glass windows and a high-speed schlieren photography system were designed and employed. Pressure rise criterion of 5%,

Table 3 – Flammability limits of methane (in vol.%) determined by various methods.

Gas	Method	Criterion	LFL	UFL
Methane	20 L sphere [15]	2% Pressure rise	4.58 ± 0.11	15.9 ± 0.3
	20 L chamber [13]	3% Pressure rise	4.9 ± 0.1	-
	120 L sphere [13]	3% Pressure rise	5.0 ± 0.1	-
	4.2 L sphere [21]	5% Pressure rise	4.6	16.0
	5.34 L chamber (this work)	5% Pressure rise	5.0 ± 0.1	16.0 ± 0.1
	14 L chamber [16]	5% Pressure rise	4.9	16.9
	5.34 L chamber (this work)	7% Pressure rise	5.1 ± 0.1	15.7 ± 0.1
	8 L chamber (Hertzberg and Cashdollar, 1983) from Ref. [13]	7% Pressure rise	5.0 ± 0.1	–
	20 L chamber [13]	7% Pressure rise	5.0 ± 0.1	15.9 ± 0.1
	20 L sphere [15]	7% Pressure rise	4.85 ± 0.11	15.1 ± 0.3
	120 L sphere [13]	7% Pressure rise	5.0 ± 0.1	15.7 ± 0.2
	25.5 m^3 sphere (Burgess et al., 1982) from Ref. [13]	7% Pressure rise	5.1 ± 0.1	–
	2.7 L chamber from Ref. [21]	10% Pressure rise	4.6	16.5
	8 L sphere [12]	Tangent criterion	4.6 ± 0.3	15.8 ± 0.4
	8 L sphere [12]	Min–max criterion	4.6 ± 0.3	15.7 ± 0.4
	12 L sphere [11]	Visual	4.9 ± 0.1	15.8 ± 0.1
	25.5 m^3 sphere (Burgess et al., 1982) from Ref. [13]	Visual	4.9	–
	Flammability porous tube with diameter of 30 mm [14]	Visual, horizontal	4.7	15.1
	Flammability porous tube with diameter of 30 mm [14]	Visual, vertical	4.7	14.9
	Flammability glass tube with diameter of 50 mm [9]	Visual, vertical[a]	5.3	14.0
	Flammability glass tube with diameter of 60 mm [15]	visual, vertical[a]	4.6 ± 0.06	16.2 ± 0.2
	Flammability glass tube with diameter of 60 mm [21]	Visual, vertical[a]	4.4	15.8
	Flammability glass tube with diameter of 60 mm [16]	Visual	4.2	16.6
	Flammability glass tube with diameter of 80 mm [16]	Visual	4.3	16.8
	ASTM E 681-01 using glass flask, V = 5 L [16]	Visual	3.8	16.9
Natural gas	5.34 L chamber (this work)	5% Pressure rise	5.0 ± 0.1	16.8 ± 0.1
	1.57 L chamber [10]	7% Pressure rise	5.0	15.6
	5.34 L chamber (this work)	7% Pressure rise	5.1 ± 0.1	16.5 ± 0.1

a Upward flame propagation.

Table 4 – Lower flammability limits of hydrogen (in vol.%) determined by various methods.

Hydrogen, H_2			
Method	Criterion	LFL	UFL
20 L chamber [13]	3% Pressure rise	5 ± 0.5	–
120 L sphere [13]	3% Pressure rise	5 ± 0.5	–
5.34 L chamber (this work)	5% Pressure rise	5.0 ± 0.1	76.5 ± 0.1
14 L chamber [16]	5% Pressure rise	4.2	77.0
5.34 L chamber (this work)	7% Pressure rise	6.0 ± 0.1	76.0 ± 0.1
8 L chamber (Hertzberg and Cashdollar, 1983) from Ref. [13]	7% Pressure rise	5 ± 0.5	76.8 ± 0.2
20 L chamber [13]	7% Pressure rise	6.0 ± 0.5	–
120 L sphere [13]	7% Pressure rise	6.5 ± 0.5	–
25.5 m^3 sphere (Burgess et al., 1982) from Ref. [13]	7% Pressure rise	7.5 ± 0.5	–
Flammability porous tube with diameter of 30 mm [14]	Visual, horizontal	4.4	–
Flammability porous tube with diameter of 30 mm [14]	Visual, vertical	4.2	–
Flammability glass tube with diameter of 50 mm [9]	Visual, vertical[a]	4.0	72.0
Flammability stainless steel tube with diameter of 50 mm [17]	Visual, vertical[a]	4.13	–
Flammability stainless steel tube with diameter of 50 mm [18]	Visual, vertical[a]	–	76
Flammability glass tube with diameter of 60 mm [16]	Visual	3.8	75.8
Flammability glass tube with diameter of 80 mm [16]	Visual	3.6	76.6
ASTM E 681-01 using glass flask, V = 5 L [16]	Visual	3.75	75.1

[a] Upward flame propagation.

recommended by EN 1839, was used to determine the flammability limits of hydrogen-enriched natural gas.

In the following sections, the experimental set-up is described at first. Then, by comparing the flammability limits of methane and hydrogen obtained in this study with the corresponding values reported in Refs. [9–18,21], the experimental system and the data analyzing method used in this study are validated. The flammability characteristics of hydrogen-enriched natural gas are discussed in detail by examining pressure rise history inside the combustion chamber together with flame propagation photos. Finally, both the lower and upper flammability limits of hydrogen-enriched natural gas are reported and compared with the corresponding values of methane–hydrogen mixtures.

2. Experimental set-up

The experimental set-up includes a constant volume combustion chamber and the systems for ignition, data acquisition and high-speed schlieren photography. The constant volume combustion chamber used in this study is a cylindrical type with the inner diameter of 180 mm and the length of 210 mm (5.34 dm^3 in volume). Two sides of the chamber are made of quartz glasses to provide optical access, allowing the observation of the combustion process taking place inside the chamber. The details of the combustion chamber are illustrated in Fig. 1. Flame propagation photos were recorded by a Redlake HG-100 K high-speed CCD camera, operating at 10,000 pictures per second.

In the combustion chamber, there are inlet/outlet valves to let fresh air in and combustion products out. The combustible mixture in the combustion chamber was prepared by adding correct amounts of fuel and air according to their corresponding partial pressures. The partial pressures were determined by initial pressure and hydrogen fraction x (the volume fraction of the hydrogen in the fuel of hydrogen-enriched natural gas or methane–hydrogen mixtures). For each test, natural gas or methane was introduced first, followed by hydrogen and then air. 2-min waiting time is required to allow the gaseous mixture inside the combustion chamber to rest. The quiescent mixture was then ignited by fusing a coiled tungsten wire, located at the center of the chamber, by applying a voltage of 24 V dc. Such an igniter releases about 10 J in 0.1 s. The pressure inside the bomb was recorded by a piezoelectric Kistler pressure transducer with a resolution of 0.01 kPa. Pressure rise criterion of 5% was used to determine the flammability limits. At near-limit conditions, at least five tests were conducted to decide whether a flame can propagate in a mixture or not.

It is worth to mention that the experimental apparatus and procedure used in this study were designed by following the new European standard EN 1839 [23], which is applicable to the measurement of the flammability limits of combustible gas or their mixtures at normal atmospheric pressure.

1. *Dimension of combustion chamber*: the internal volume of the combustion chamber is larger than 5 dm^3, which satisfies the requirement of EN 1839.
2. *Ignition system*: in this study, the method of fusing tungsten wire was used to ignite the quiescent fuel–air mixture, while EN 1839 recommends using nichrome wire. This is because Takahashi et al. [24] found that fusing a nichrome wire is not very suitable for the flammability limits measurement and recommended using metals with high melting point (such as tungsten). Similar ignition system was also used in Refs. [19–21].
3. *The pressure rise criterion*: 5% pressure rise criterion, recommended by EN 1839, was used in this study, meaning that an ignition will be regarded as a successful one if the ignition is followed by a pressure rise of at least 5% of the initial pressure inside the combustion chamber before igniting.

In this study, hydrogen and methane with purity of 99.995% and 99.99% respectively were used. The constitution of natural gas is listed in Table 2. The test mixtures were prepared with the maximum uncertainty of 0.1 vol.% for the mole fraction of each component.

3. Experimental system validation

To validate the experimental set-up as well as the data analysis method used in this study, both the lower flammability limit (LFL) and the upper flammability limit (UFL) of methane and hydrogen were measured and compared with the corresponding flammability data reported in Refs. [9–18,21]. Tables 3 and 4 summarize the flammability limits of methane and hydrogen obtained by using various methods and criterions respectively.

Table 3 shows that the lower flammability limit of methane lies in the range between 3.8% and 5.3%, while the upper flammability limit between 14.0% and 16.9%. It is worth to mention that all these 'boundary values' were obtained by using flammability tubes or flasks made of glass. For the flammability limits measured by using combustion chambers, the variation is relatively small, especially when the internal volume of the chamber is larger than 5 dm^3.

Fig. 2 summarizes the experimental results obtained by using constant volume combustion chambers (or explosive vessels) with different internal volumes. From Fig. 2a and b, we can see that when the internal volume is less than 5 dm^3 (first two bars from left, reported in [21]), the flammability limits differ greatly with these obtained by using combustion chambers whose internal volumes are larger than 5 dm^3 (present study, [12,13,15,16]). Our experimental results agree well with the majority of the reported values (obtained by using spherical or cylindrical chambers with internal volumes varying from 8 dm^3 up to 25.5 m^3). As such we conclude that our results agree well with these reported values and this proves the reliability of the experimental system used in this study.

For hydrogen (as summarized in Table 4), the lower flammability limit is 3.6–4.4% when using flammability tubes, while most of the lower flammability limit measured by combustion chamber is greater than 5%. By plotting the experimental results of LFL obtained by using combustion chambers, Fig. 3a shows that both internal volume of combustion chamber and pressure rise criterion have great effects on the lower flammability limit of hydrogen. Our results agree well with the value reported in Ref. [13]. An exceptionally low LFL value was reported in Ref. [16]. Fig. 3b shows that the upper flammability limit of hydrogen is not sensitive to the design of the combustion chamber as well as the pressure rise criterion used. And the UFL of hydrogen measured in this study agrees well with all the reported ones.

The effects of combustion chamber dimension and shape were studied by Takahashi et al. [25] by using a series of combustion chambers. They concluded that for cylindrical chambers, the diameter should be at least 30 cm and the height at least 60 cm, which gives the minimum internal

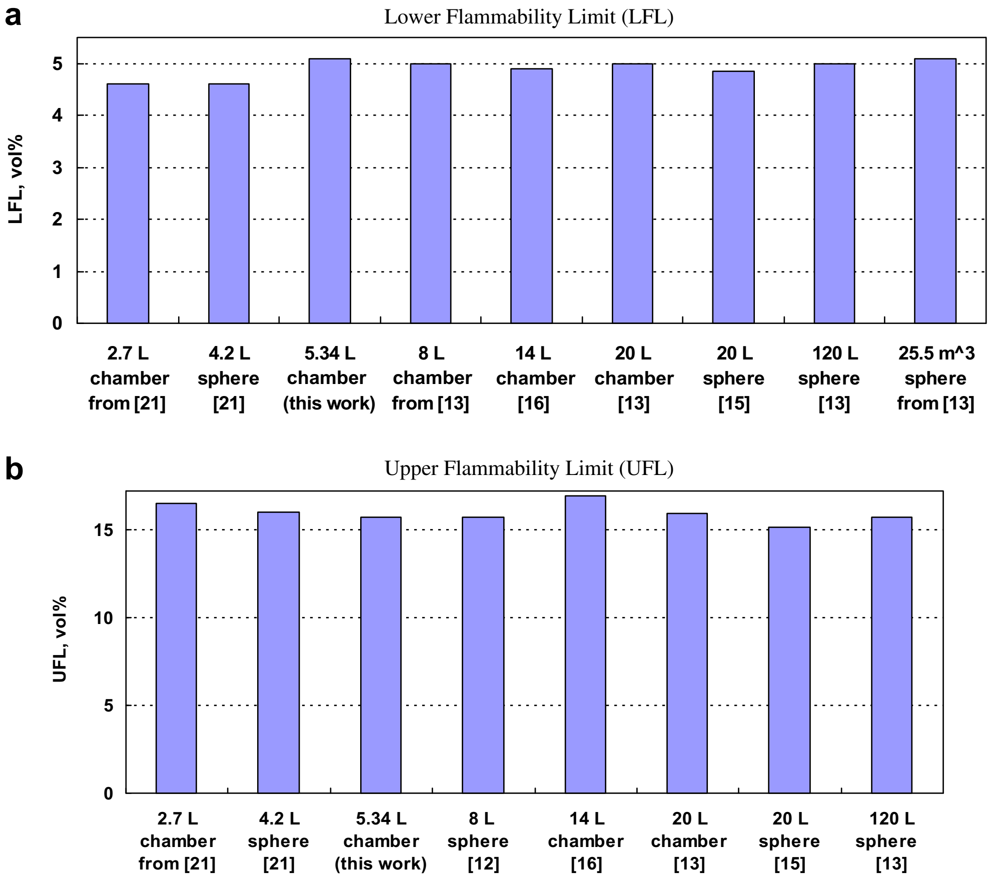

Fig. 2 – Flammability limits of methane measured by using combustion chambers with different volumes.

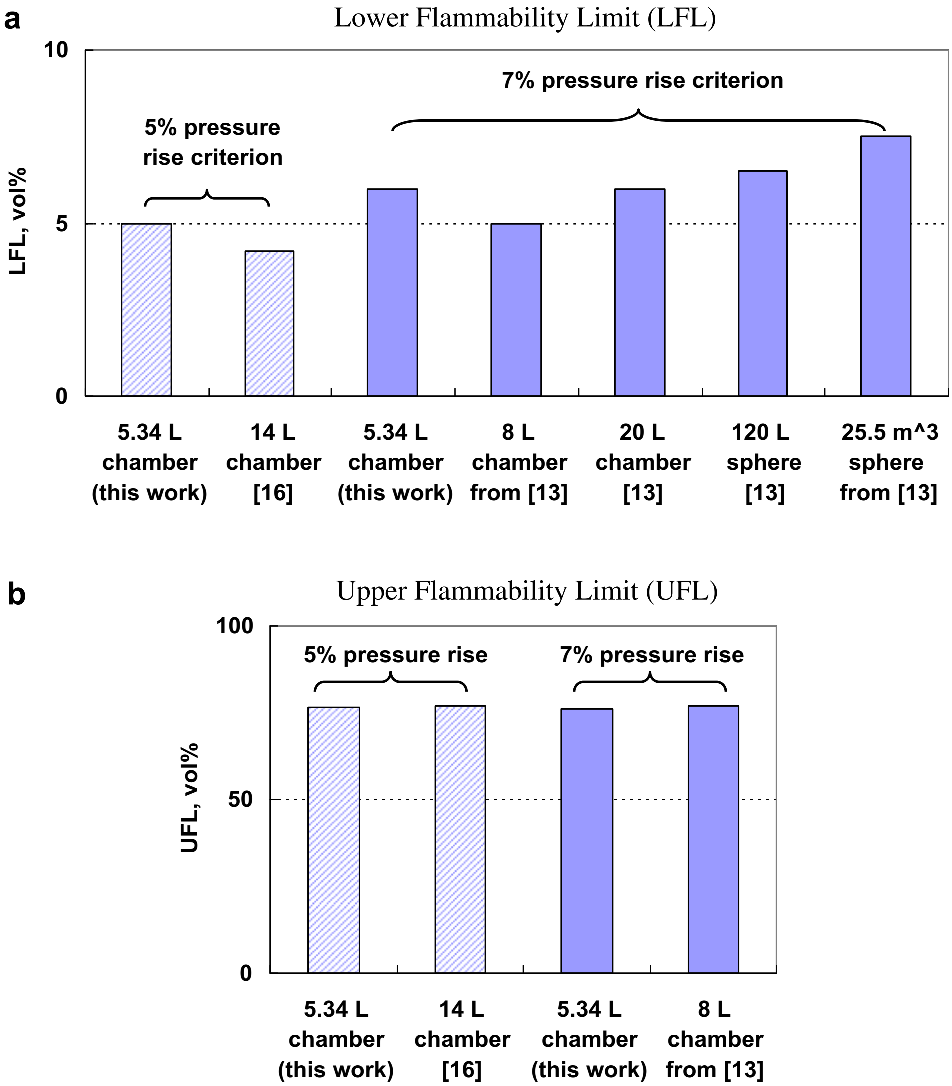

Fig. 3 – Flammability limits of hydrogen measured by using combustion chambers with different volumes.

volume of 42 dm^3. Our recent research experience showed that by using a much smaller combustion chamber (5.34 dm^3), it is possible to achieve the experimental condition similar to that of a very large chamber (25.5 m^3 [13]). In fact, our results agree well with these obtained in Ref. [13]. Therefore, this study provides evidence that the requirement of EN 1893 on the minimum internal volume of a close chamber or vessel (5 dm^3) is appropriate.

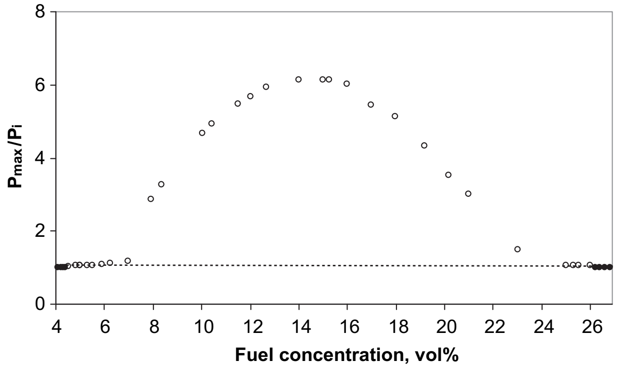

Fig. 4 – Determination of the flammability limits of hydrogen-enriched natural gas with hydrogen fraction of 40%, where hollow points represent successful attempts and solid points for these unsuccessful ones.

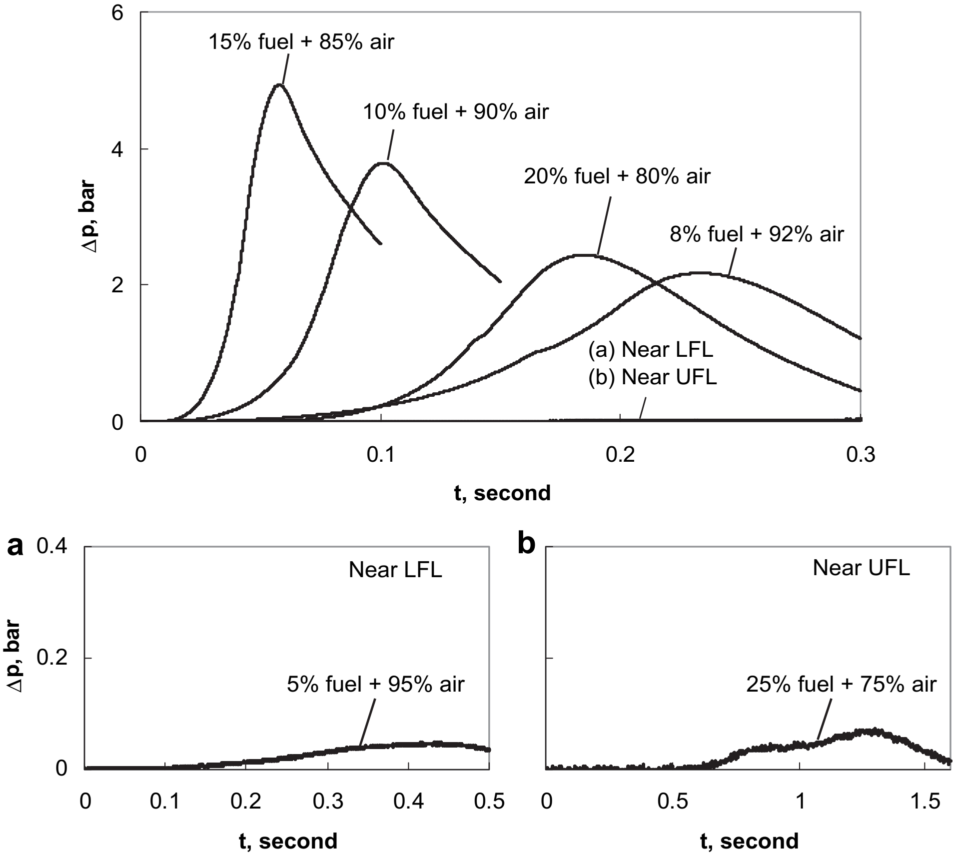

Fig. 5 – Pressure rise inside the combustion chamber after igniting the quiescent hydrogen-enriched natural gas (the hydrogen fraction of the fuel is 40%) and air mixture. (a) and (b) show zoomed pressure rise history inside the combustion chamber for the fuel–air mixtures that are very close to the lower flammability limit (LFL) and the upper flammability limit (UFL) respectively.

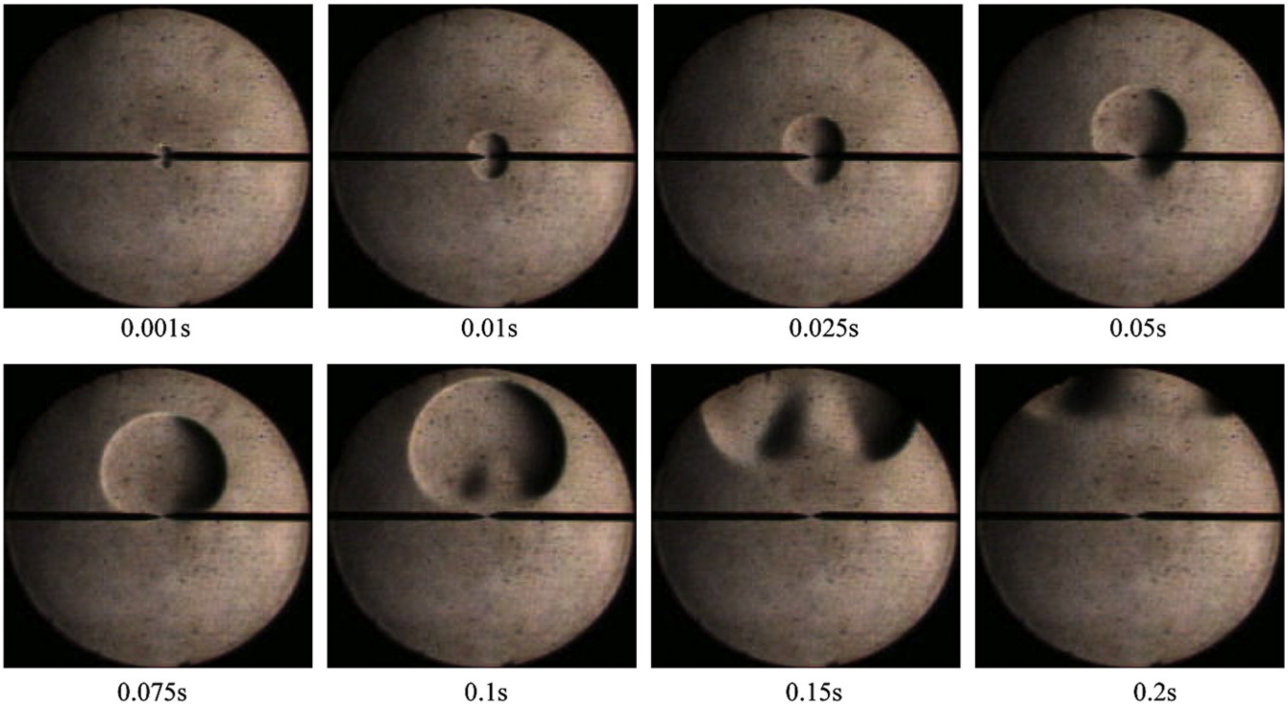

Fig. 6 – Flame development of hydrogen-enriched natural gas and air mixture near its lean limit (5% fuel + 95% air; the hydrogen fraction of the fuel is 40%).

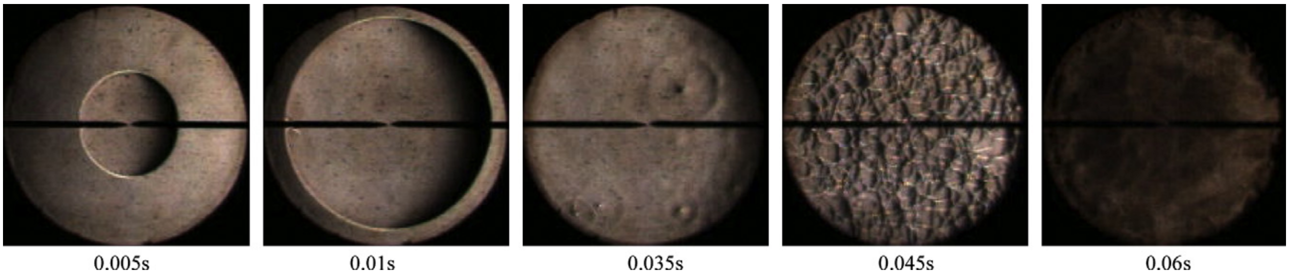

Fig. 7 – **Flame development of hydrogen-enriched natural gas and air mixture near its stoichiometric condition (15% fuel + 85% air; the hydrogen fraction of the fuel is 40%).**

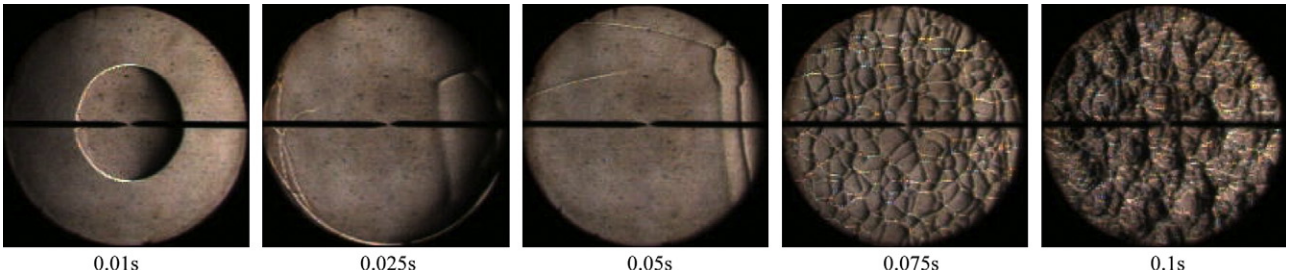

Fig. 8 – **Flame development of hydrogen-enriched natural gas and air mixture in between its lean limit and stoichiometric conditions (10% fuel + 90% air; the hydrogen fraction of the fuel is 40%).**

4. Flammability limits of hydrogen-enriched natural gas

To obtain the flammability limits of hydrogen-enriched natural gas, we tested not only near-limit mixtures with air, but also mixtures in between them, by gradually increasing the concentration of hydrogen-enriched natural gas from its lean limit to rich limit. Fig. 4 illustrates how to measure the flammability limits and provides information on the range of the combustible mixtures that were tested during the experiments, using hydrogen-enriched natural gas with hydrogen fraction of 40% as an example. It was observed that the ratio of the peak pressure inside the combustion chamber to its initial pressure is very close to 1 for near lean-limit mixtures (fuel concentration around 4% to 6%). With the increase of the fuel concentration, the ratio increases until it reaches the maximum value of 6 near the stoichiometric condition; then it starts to drop with the further increase of the fuel concentration. And when the fuel concentration is greater than 25%, it has a value near 1 again. By employing pressure rise criterion of 5%, the lower and upper flammability limits of the hydrogen-enriched natural gas whose constitution is 40% hydrogen and 60% natural gas are determined as 4.8% and 26.0% respectively.

In this study, both the pressure rise history (as shown in Fig. 5) and the corresponding flame photos captured by the high-speed schlieren photography system are available and therefore make it possible for us to study detailed combustion processes inside the combustion chamber. We found that:

- The pressure inside the chamber rises slowly at near lean-limit (Fig. 5a) and near rich-limit conditions (Fig. 5b). Using near lean-limit condition as an example, Fig. 5a shows that the pressure rise is very limited (about 0.05 bar) and this is typical for the combustion of a combustible mixture on the edge of its flammability limits. Schlieren photos (see Fig. 6) show that at the early stage of the combustion, spherical flame propagation is observed. The flame front expands outwardly because of the slow but continuously combustion processes on it. At the later stage, the flame starts to rise and changes its shape, similar to the process when an air bubble rises in water. It is the density difference that drives the flame upward because the combustion products are hotter than the surrounding gases.

Table 5 – **Flammability limits of hydrogen-enriched natural gas.**

Hydrogen-enriched natural gas						
Hydrogen fraction (%)	0	20	40	60	80	100
LFL (vol.%)	5.0	5.0	4.8	5.0	5.0	5.0
UFL (vol.%)	16.8	20.9	26.0	33.9	47.0	76.5

Table 6 – **Flammability limits of hydrogen-enriched methane.**

Hydrogen-enriched methane						
Hydrogen fraction (%)	0	20	40	60	80	100
LFL (vol.%)	5.0	4.6	4.4	4.4	4.6	5.0
UFL (vol.%)	16.0	19.9	26.0	33.5	47.6	76.5

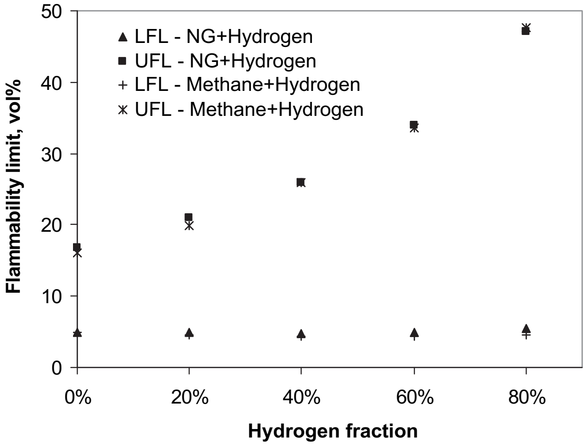

Fig. 9 – Comparison of flammability limits of hydrogen-enriched natural gas (NG) and methane–hydrogen mixtures.

- The maximum pressure rise (about 5.5 bar) happens when the fuel concentration is 15%, which is near the stoichiometric condition (see Fig. 5). It takes 0.06 s for the pressure inside the chamber to reach its peak value. Central-located spherical flame was observed when examining the schlieren photos (given in Fig. 7), meaning that the flame expands at all directions with same speed. After the flame front reaches the chamber wall, the smoothness of the flame front starts to break and well-developed flaws can be observed at 0.45 s after ignition, showing that there exist active chemical reactions. At the time when the pressure inside the chamber reaches its peak value (t = 0.06 s), flame photos showed that the combustion is already at its decayed phase.
- For mixtures in between flammability limits and stoichiometric conditions: as shown in Fig. 5, the nearer a mixture is to the stoichiometric condition, the faster the pressure rises to a higher peak value. Fig. 8 shows typical combustion processes for these mixtures, using the fuel/air mixture with 90% air as an example.

To obtain the flammability limits of hydrogen-enriched natural gas, the processes mentioned above need to be repeated when its hydrogen fraction is changed. In this study, both the lower and the upper flammability limits of hydrogen-enriched natural gas with hydrogen fraction of 20%, 40%, 60%, and 80% respectively as well as these of natural gas and pure hydrogen were obtained experimentally and are summarized in Table 5.

Moreover, the flammability limits of hydrogen and methane mixtures with hydrogen fraction of 20%, 40%, 60%, and 80% respectively were measured to answer the question whether the flammability data of methane–hydrogen mixtures are good enough to represent these of hydrogen-enriched natural gas. The experimental results are given in Table 6.

The effect of hydrogen fraction on the flammability limits of hydrogen-enriched natural gas is clearly illustrated in Fig. 9. With the increase of the hydrogen fraction, the upper flammability limit increases; this means that the flammable range of hydrogen-enriched natural gas and air mixtures is extended when there is more hydrogen inside the fuel. But, changing the hydrogen fraction has limited influence on the value of the lower flammability limit. Similar trends were also found in the flammability data of methane–hydrogen mixed fuels (see Table 6).

By comparing the flammability limits of hydrogen-enriched natural gas and the corresponding values of methane–hydrogen

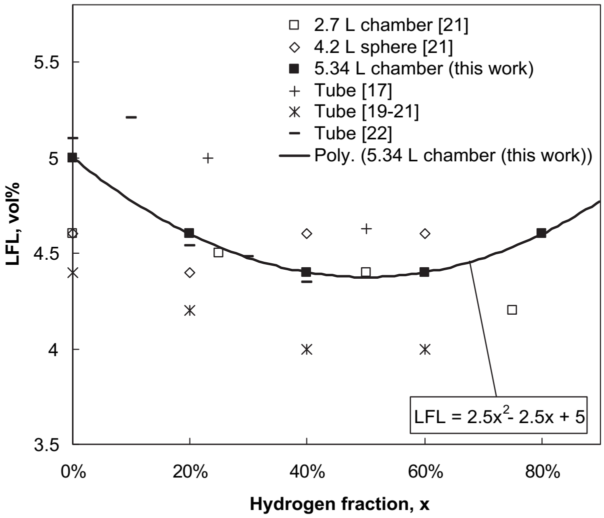

Fig. 10 – Lower flammability limits of methane–hydrogen mixtures.

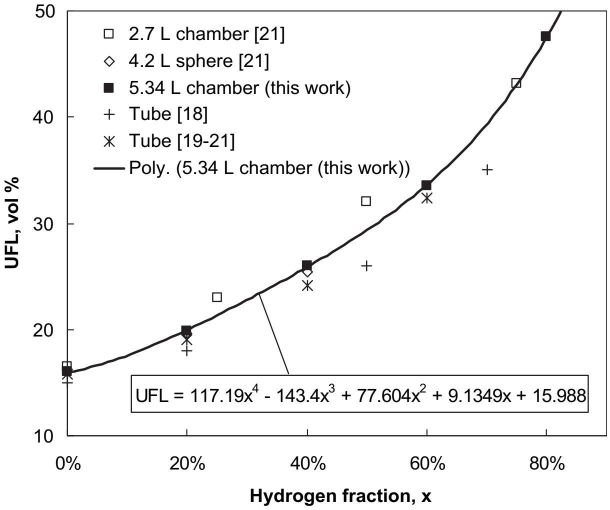

Fig. 11 – Upper flammability limits of methane–hydrogen mixtures.

mixtures in Fig. 9, it can be seen that the two sets of the flammability data are very close to each other. Our experimental results show that the flammability data of methane–hydrogen mixtures can be applied to hydrogen-enriched natural gas as long as their hydrogen fractions are the same.

It is worth to remark that the flammability limits measured by using flammability tubes and combustion chambers are sometimes different. Figs. 10 and 11 plot the available lower and upper flammability limits of methane–hydrogen mixtures respectively. Fig. 10 shows that the measurement instrument has great effects on the lower flammability limit results, while the upper flammability limit measured by different methods tends to agree with each other (see Fig. 11). Therefore, the understanding of the experimental details (such as what kind of the device was used, how the flame was ignited and which criterion was applied) is essential for the proper usage of any flammability data.

Finally, the correlations for the lower and upper flammability limits of methane–hydrogen mixtures are obtained based on the experimental results of this study.

$$\mathrm{LFL} = 2.5x^2 - 2.5x + 5 \tag{1}$$

$$\mathrm{UFL} = 117.19x^4 - 143.4x^3 + 77.604x^2 + 9.1349x + 15.988 \tag{2}$$

In these correlations, x is the volumetric fraction of hydrogen in a methane–hydrogen mixture. By plotting the LFL and UFL predicted by the correlations together with the experimental data (as shown in Figs. 10 and 11), it can be seen that the correlations can represent the experimental data with reasonable accuracy. The standard deviations S_N of these correlations are summarized in Table 7, which are defined as below:

Table 7 – Standard deviations of experimental and calculated flammability limits data.

Standard deviation						
Hydrogen fraction (%)	0	20	40	60	80	100
For LFL (vol.%)	0.0	0.33	0.33	0.49	0.33	0.0
For UFL (vol.%)	0.66	0.79	0.10	0.29	0.47	0.01

$$S_N = \sqrt{\frac{1}{N}\sum_{i=1}^{N}(x_i - \bar{x})^2} \tag{3}$$

where $N = 3$; x_1, x_2 and x_3 stand for the flammability limits of hydrogen-enriched natural gas (measured in this study), methane–hydrogen mixtures (measured in this study) and the corresponding values calculated by using the correlations (Eqs. (1) and (2)) respectively; and $\bar{x}$ is the mean value of x_1, x_2 and x_3.

5. Conclusion

The flammability limits of hydrogen-enriched natural gas with various hydrogen fractions ranging from 0% to 100% were measured by employing a combustion chamber together with a high-speed schlieren photographic system. Combustible mixtures were ignited by fusing a coiled tungsten wire located at the center of the combustion chamber. Both the lower flammability limit (LFL) and the upper flammability limit (UFL) of hydrogen-enriched natural gas were obtained. Experimental results showed that the flammability data of methane–hydrogen mixtures are applicable to hydrogen-enriched natural gas. Future studies will be conducted to investigate the effect of natural gas constitute on the flammability limits of hydrogen-enriched natural gas.

This paper also reviewed the flammability data of methane–hydrogen mixtures available in the literature with remarks that experimental methods do have influence on the flammability limit data, especially for the lower flammability limit of hydrogen. Therefore, special attention needs to be given to the method as well as the criterion for flammability data before using them.

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