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Experimental study of lean flammability limits of methane/hydrogen/air mixtures in tubes of different diameters

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ABSTRACT

Lean limit flames in methane/hydrogen/air mixtures propagating in tubes of internal diameters (ID) of 6.0, 8.9, 12.3, 18.4, 25.2, 35.0, and 50.2 mm have been experimentally studied. The flames propagated upward from the open bottom end of the tube to the closed upper end. The content of hydrogen in the fuel gas has been varied in the range 0–40 mol%. Lean flammability limits have been determined; flame shapes recorded and the visible speed of flame propagation measured. Most of the observed limit flames in tubes with diameters in the range of 8.9–18.4 mm had enclosed shape, and could be characterized as distorted or spherical flame balls. The tendency was observed for mixtures with higher hydrogen content to form smaller size, more uniform flame balls in a wider range of tube diameters. At hydrogen content of 20% or more in the fuel gas, limit flames in largest diameters (35.0 mm and 50.2 mm ID) tubes had small, compared to the tube diameter, size and were "lens"-shaped. "Regular" open-front lean limit flames were observed only for the smallest diameters (6.0 mm and 8.9 mm) and largest diameters (35.0 and 50.2 mm ID), and only for methane/air and (90% CH_4 + 10% H_2)/air mixtures, except for 6 mm ID tube in which all limit flames had open front. In all experiments, except for the lean limit flames in methane/air and (90% CH_4 + 10% H_2)/air mixtures in the 8.9 mm ID tube, and all limit flames in 6.0 mm ID tube, visible flame speeds very weakly depended on the hydrogen content in the fuel gas and were close to- or below the theoretical estimate of the speed of a rising hot bubble. This observation suggests that the buoyancy is the major factor which determines the visible flame speed for studied limit flames, except that last mentioned. A decrease of the lean flammability limit value with decreasing the tube diameter was observed for methane/air and (90% CH_4 + 10% H_2)/air mixtures for tubes having internal diameters in the range of 18.4–50.2 mm. This effect has been attributed to the stronger combined effect of the preferential diffusion and flame stretch in narrower tubes for flames which resemble rising bubble.

1. Introduction

Experimental studies on flammability limits and limit flames in combustible mixtures are important both from a fundamental and practical points of view. Knowledge of flammability limits is essential for safety standards and in the design of lean-combustion devices. On the other hand, new knowledge can be obtained, or theoretical predictions tested in such studies on flame stretch and preferential diffusion effects on flame propagation and extinction in non-unity Lewis number mixtures. Practical flammability tests are performed at normal gravity, in tubes or closed vessels. One of the standard flammability tests utilizes a so-called standard flammability tube: a 50 mm diameter and 1800 mm long tube open from the bottom end and closed at the top end [1]. In this test, the mixture is considered to be flammable if, after the ignition near the bottom end, the flame propagates upward throughout the entire tube length.

The flammability tests used in the present work, as described below, are similar to this standard one. The available experimental information on the effect of the tube diameter on flammability limits, obtained in test conditions similar to ones used in the present work is rather limited. Tests of this kind have been performed in tubes of different diameters in [1–4]. Practically no difference in lean flammability limits (LFL) for hydrogen/air mixtures (Lewis number, $Le < 1$) in tubes with 25 mm and 53 mm inner diameter (ID) was found in [1]. For methane/air mixtures, authors of [1] reported narrower LFL ($Le < 1$) for the smaller diameter tube (5.24 mol% of methane in 50 mm ID tube against 5.48 mol% in 25 mm ID tube). At the same time, the flammability tests performed in [2] for methane/air mixtures in similar diameters tubes

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Nomenclature

A	constant
g	gravity acceleration (m/s^2)
R	tube radius (m)
S_b	speed of an open rising bubble in a tube (m/s)

Greek symbols

Θ	gas thermal expansion ratio
φ	equivalence ratio

showed the opposite trend: leaner mixture was able to support flame propagation in the narrower tube (5.1 mol% of methane in 50 mm ID tube against 4.9 mol% in 24 mm ID tube). This discrepancy can be explained by the sensitivity of the flammability limit to the source of ignition [2,3]. Wider flammability limits were also observed in narrower tubes for propane/air mixtures on the rich side ($Le < 1$) by the authors of [4]. The effect of the tube diameter on the pressure limit of flame propagation for propane/air mixtures was experimentally studied in [3]. Authors carried out experiments in various-diameter tubes (16–66 mm) with closed upper end and bottom end connected to a large-volume ignition chamber. The mixture was considered inflammable if flame did not pass through the bottom tube mouth. A systematic increase of minimum pressure for the flame propagation with the decrease of the tube diameter was reported for lean flames ($Le > 1$). For fuel-rich propane/air flames ($Le < 1$), measured pressure limits versus tube diameter varied irregularly, which was attributed by the authors to irregular instabilities of fuel-rich flames in the ignition chamber [3]. Though the above cited experimental results are rather inconsistent, some of them indicate that tube diameter may significantly affect flammability limits in mixtures with non-unity Lewis number. The inconsistency could possibly result from using, in some works, of an inappropriate ignition source, which could not launch a flame in mixtures potentially capable to sustain flame propagation in a tube.

In many cases, lean limit flames closely resemble a rising low density open bubble in a tube filled with a fluid with higher density. The shape and visible speed of such flames are similar to ones theoretically predicted for such a rising bubble [5,6]. It was suggested that the observed in some experiments widening of the flammability limits in $Le < 1$ mixtures in narrower tubes for such flames can be explained by the coupled flame stretch and preferential diffusion effect [2]. The positive flame stretch is produced by the divergent gas flow around a rising open bubble of hot combustion product gases. The estimations yielded higher maximum stretch rates for smaller tube diameters for such flames. Because the combined positive flame stretch and preferential diffusion effect increases flame temperature in mixtures with Lewis number, $Le < 1$ [7], one can expect that LFL in smaller diameter tubes will be wider for such mixtures (unless the tube becomes so narrow that heat losses to the tube wall become a dominant factor affecting the flammability limit).

In general, the shape of a limit flame propagating in a tube may not resemble an open rising bubble. For example, enclosed flames having droplet-like shape were described in [3] for limit flames in rich propane-air mixtures ($Le < 1$) propagating upward in a 50.8 mm diameter tube at reduced pressures. A globular flame traveling with a nearly constant speed was observed by authors of [1] for methane/air mixture near LFL in a 50.8 mm ID tube. Besides, some flammability tests involve only earlier stages of the flame propagation in a tube, for which stretch rate estimations based on mechanics of a steadily propagating rising bubble may not be applicable. Nevertheless, it is understood that preferential diffusion effects may play an essential role in all flammability experiments in which the flame is not flat and divergence of the gas flow is present, in particular when flames are affected by the buoyancy. An indicative of the importance of preferential diffusion effect is significant difference in flammability limits determined at different conditions for methane/air mixtures and methane/hydrogen/air mixtures containing 20, 40, and 60 mol% of hydrogen in the fuel gas in [8]. Flammability limits in that work were determined experimentally using two different criteria: partial, more that 10 cm, flame propagation in a 60 mm diameter 300 mm long tube (German standard DIN 51649) and 5% pressure rise threshold in a 200 mm diameter enclosed spherical vessel. In addition, flammability limits in [8] were determined numerically for two configurations: a flat and spherical flame. The discrepancy between LFL found by different methods strongly increased with the increase of hydrogen concentration in the tested mixture. It is known that the "sensitivity" of the mixture to preferential diffusion effects is mainly determined by the value of the parameter $(1-Le)$ [7]. The average value of this parameter for lean methane/hydrogen/air mixtures strongly increases with the increase of the hydrogen content in the mixture, which allows to suggest that the observed trends are related to flame stretch/preferential diffusion effects.

The aim of this work is to experimentally investigate the effect of the tube diameter on lean flammability limits, limit flame shapes and visible propagation speeds for methane/hydrogen/air mixtures. Upward flame propagation in tubes closed at the upper end and opened at the bottom end was studied. Similar to the standard tube methodology [1], reaching of the upper tube end by the flame was considered as a criterion of flammability. A special care was taken to provide a maximally reliable ignition of the tested mixtures, so that measured flammability limits could be related only to the ability of the mixture to support the flame propagation in a tube, but not to the method of ignition. Methane/hydrogen/air mixtures were selected because they allow for strong variation of the "strength" of the preferential diffusion effects through the variation of the hydrogen content. Besides, mixtures of natural gas with hydrogen are expected to be of great practical importance in the near future, especially for lean-combustion devices with reduced emission of harmfully species. The widest tube used in the experiments had a size of a standard flammability tube, thus flammability limits determined in that tube can be taken as "standard" ones for safety issues.

2. Experimental methodology

Cylinder gases (technical methane and hydrogen of better than 99.9% purity) were used in the experiments. Fuel gases were mixed with dried house air. The tested fuel gases were methane and methane/hydrogen mixtures containing 10%, 20%, 30%, and 40% of hydrogen. The mixtures of fuel gas with air will be referred to in the following text as methane/air, and, by hydrogen molar content in the fuel, as 10% H_2, 20% H_2, 30% H_2, and 40% H_2 mixtures.

Fig. 1 shows a schematic of the experimental setup used to study the limit flames. Pyrex tubes with an internal diameter (ID) of 6.0, 8.9, 12.3, 18.4, 25.2, 35.0, and 50.2 mm were used in the experiments. The 6.0 mm diameter was 450 mm long, tubes of 8.9 mm, 12.3 mm, and 18.4 mm ID were 900 mm long, and tubes of 25.2 mm, 35.0 mm, and 50.2 mm ID were 1700 mm long. The