STABILISATION OF LAMINAR INVERTED ULTRA-LEAN HYDROGEN-METHANE-AIR FLAMES

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Abstract

Blowoff characteristics of inverted flames propagating in ultra-lean hydrogen-metane-air mixtures and stabilised at the end of a 2 mm diameter rod, have been studied experimentally for different hydrogen contents in the fuel gas blend. It was found that flames with hydrogen content in the fuel gas of 60% and more can be stabilsed at equivalence ratios well below the theoretical lean flammability limit for the corrsponding planar flames. Anomalous blowoff behavior was observed for flames in mixtures with a hydrogen content of 40% and more in the fuel gas. The blowoff of these flames occurred at a decreased flow velocity, after some critical value of the velocity was achieved. Below this value, the flame could not be stabilised. The stabilisation rod temperature, near its trailing edge, was measured with an infrared pyrometer. Low values of the measured rod temperature suggest that the flames studied were stabilised by flame stretch effects, rather than due to the heat losses to the stabilisation rod. The experimentally observed anomalous blowoff behaviour in mixtures with high hydrogen content in the fuel gas is attributed to strong combined flame stretch/preferential diffusion effects.

Introduction

Inverted flames stabilised at the end of a thin rod or at the edge of a thin plate are often used in studies of premixed flame stabilisation limits. The symmetry of such flames and the absence of the influence of the surrounding atmosphere may facilitate modelling and understanding of the mechanisms of flame stabilisation. Besides, from stabilisation viewpoint, such flames are likely to be physically similar to practically relevant flames on multi-slit or multi-hole burners.

Flame stabilisation occurs when the burning velocity at the flame base is balanced by the local mixture flow velocity. In early studies of inverted flame stabilisation [1, 2], the authors assumed that such a balance was achieved due to the heat losses from the flame to the rod or the plate edge. According to the suggested mechanism, if the flame was displaced from its stable location towards the stabilisation edge, its speed would decrease and the flame would move back to its original location, and vice versa. Flame blowoff was related by authors to the increase of the heat flux from the reaction zone to the unburned mixture due to the positive flame stretch rate [1, 2]. According to this hypothesis, when the mixture flow velocity was increased, the flame temperature decreased, eventually causing local flame extinction near the flame anchoring location. The flame stretch, however, affects the diffusion transport rate as well as heat transport rate [3]. The combined effect leads to a lowering of the flame temperature by the positive flame stretch only for mixtures with Lewis number, Le > 1, while in Le < 1 flames, the effect is opposite.

In later works [4, 5], estimates based on experimental measurements showed that heat losses to the stabilisation rod or plate may be negligible near the blowoff limit. Analytical [5] and numerical [6] investigations demonstrated that adiabatic inverted fames can be stabilised due to flame stretch effects only. Mechanisms of the flame stabilisation suggested by the authors of [5, 6] are relevant, however, only to flames in which the burning velocity decreases with increasing positive stretch rate according to the equation:

$$S_L = S_L^o - LK, \tag{1}$$

where S_L is the laminar burning velocity of the stretched flame, S_L^o is the laminar burning velocity of the planar zero-stretch flame, L is the measure of the response of the flame to stretch, referred to as the Markstein length [7], and K is the flame stretch rate.

The sign of the Markstein length, L, depends on the value of the mixture Lewis number, and becomes negative for a sufficiently small Lewis number. Some small Lewis number flames, which may exhibit such behaviour, are of practical importance. In particular, lean flames of hydrogen-containing blends may have negative Markstein length. Indeed, numerical simulations yield negative Markstein lengths for sufficiently lean hydrogen-methane-air mixtures at high hydrogen content in the fuel gas mixture [8, 9]. Hydrogen-containing fuels, such as syngas or hydrogen-methane blends, may become dominating practical fuels in the near future. Increasing requirements for CO_2 and CO emission reduction stimulate the development of industrial technologies for syngas synthesis which exploit the water shift reaction ($CO + H_2O \rightarrow CO_2 + H_2$) to reduce the CO content leading to an increasing hydrogen content in the syngas. Hydrogen becomes the major fuel component in syngases, conditioned in such a way.

From a practical point of view, there is an increasing interest in ultra lean combustion of hydrogen containing blends. Burning ultra-lean flames in practical devices may increase their efficiency and provide low NO_x and CO emission levels.

The objective of this work is to experimentally study the blowoff behaviour of laminar inverted Bunsen flames propagating in hydrogen-methane-air mixtures at different hydrogen contents. Such studies may provide a better insight on the general tendencies and limitations related to hydrogen-containing ultra lean flame stabilisation, which may be useful for designing practical burners operating on such mixtures. At the same time, the reported experimental results may stimulate theoretical investigations of stabilisation mechanisms of such flames.

Experimental setup

The setup used in the current work is schematically shown in Fig.1. The setup was similar to the one used by Lewis and Elbe [1] for inverted Bunsen flames. The flames were stabilised above the top end of a vertical cylindrical copper rod of 2.0 mm diameter, inserted in a 35 cm long Pyrex tube of 12.4 mm internal diameter. The bottom end of the rod was mounted on a positioning system, which provided angular and X-Y positioning, and facilitated thereby the alignment of the rod inside the tube. The rod was passed through a rubber stopper with a small central hole, so that the rubber stopper was located near the bottom end of the rod. A mixture was fed through the side pipe of a metal T-shaped tube. The straight section of the T-tube, the diameter of which matched the Pyrex tube diameter, was air-tightly connected with the Pyrex tube. A segment of a soft silicone hose, one end of which was pulled on the bottom end of the T-tube and the other end - on the rubber stopper, was used to mechanically uncouple the rod and the tube. The positioning system and the Pyrex tube were mounted on separate holders. A strip of a metal knitted fabric was rolled onto the rod and filled the space between the rod and the wall of the T-tube above the mixture inlet. This was done to homogenize the mixture flow and to prevent flame penetration into the feeding system in the case of flashback. The top end of the rod was about 0.5 mm above the upper edge of the tube.

Hydrogen-methane-air mixtures were prepared in-line using three calibrated mass flow controllers (MFCs). A wide dynamic range of gas flow velocities had to be used in the experiments. At the same time, the accuracy of MFCs becomes small at small gas flow rates. To provide a better accuracy for the composition of the tested mixtures, the gas flow through the MFCs was kept close to the maximum, limited for different mixture compositions either by the air- or the hydrogen MFC available range. A fraction of the total mixture flow was directed to the Pyrex tube though a ball rotameter, which measured the mixture flow rate through the tube. The rest of the mixture flowed thought a separate pipe to a ventilation system. The mixture flow through the tube was regulated using two needle valves, one installed in the exhaust line and another one before the rotameter. Two rotameters, with measuring ranges ratio of 5:1, were used to cover the whole required range of the flow rate variations.

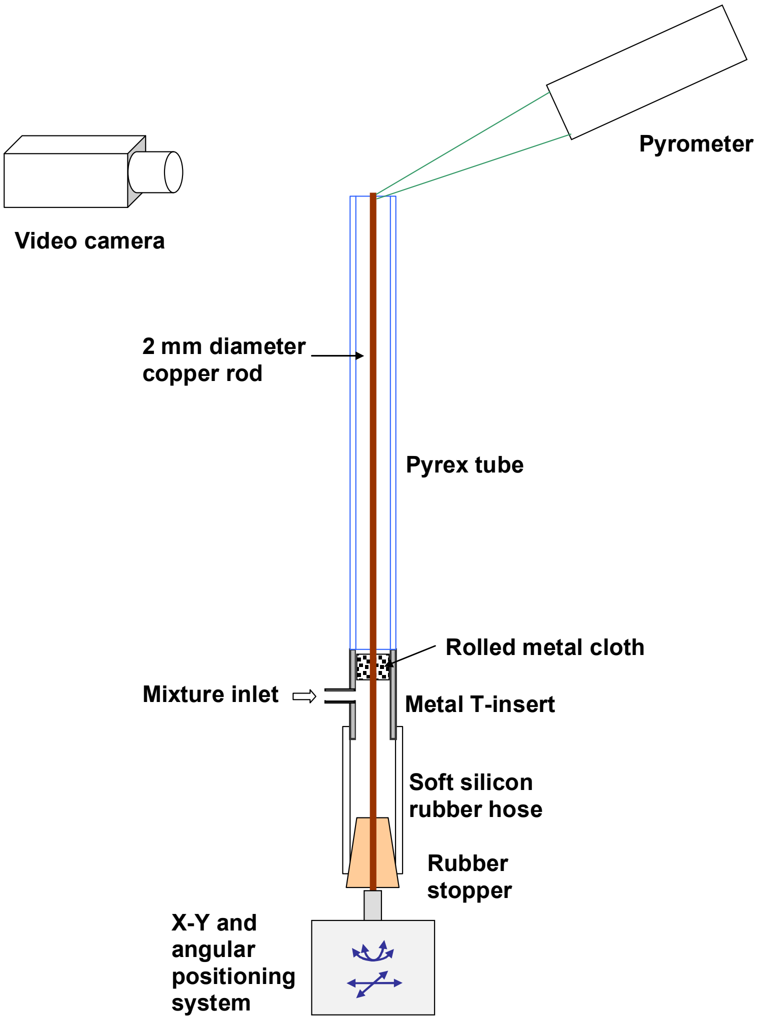


Figure 1. Schematic of the experimental setup.

An AVT PIKE F-032B BW video camera was used to register flame shapes and for real time visualisation of the hydrogen-air flames. The temperature of the stabilisation rod was measured using a small-spot infrared pyrometer PYROSPOT 40L, operating in the wavelength region of 8-14 mm. The measuring spot was focused onto the side surface of the rod and centred ~1.5 mm below the top edge of the rod. The pyrometer was installed at ~20°

angle to the horizon, to avoid vignetting of the sampled light by the tube wall. For the temperature measurements, the rod side surface at the location of the measuring spot was coated with a thin layer of black paint to increase the rod emissivity.

One of two different strategies was used to measure the stabilisation limits, depending on the slope of the experimental curves which define these limits in the coordinates "flow velocity against equivalence ratio". When such an experimental curve was nearly horizontal, the mixture flow velocity through the Pyrex tube was eventually changed, after the flame had been stabilised, until the flame stabilisation limit was reached. The mixture composition was kept constant in this kind of measurements. In the case when experimental stabilisation curve was nearly vertical, the limit of flame stabilisation was approached by the eventual decreasing of the equivalence ratio at a constant flow rate. At intermediate slopes, the first strategy was used in most cases, though some measurements were reproduced with the second method, for the control purposes.

The tested fuel gases were pure methane, pure hydrogen and hydrogen-methane mixtures with a 20%, 25%, 40%, 60%, and 80% hydrogen volume fraction. In the text below, the fuel mixtures and corresponding flames will be identified by the hydrogen volumetric content in the fuel gas, e.g. a $0.2H_2$ mixture refers to a mixture of the ($20\%H_2 + 80\%CH_4$) in the fuel gas blend with air. The maximum flow velocities in the experiments were limited by the value 4.5 m/s.

Results and discussion

Figure 2 shows the measured stabilisation limits for methane-air, $0.2H_2$ and $0.25H_2$ mixtures, in the coordinates "average mixture velocity inside the tube against total equivalence ratio". Upper branches of the stabilisation curves correspond to blow-off limits, and lower, small-velocity branches correspond to the penetration limits. The right- most points on the lower braches correspond to the maximum equivalence ratio at which flashback did not yet occur.

For the methane-air flames, the observed blow-off behaviour is in agreement with earlier results by Lewis and Elbe [1]. For mixtures, in which a stable flame can be achieved, blowoff occurs when the flow velocity is eventually increased. Higher velocities are required for the flame blowoff at higher equivalence ratios. Only flames with equivalence ratios well above the theoretical lean flammability limit (LFL) for the planar 1-D methane-air flame ($\varphi = 0.49$, see Table 1) could be stabilised.

Flames in $0.2H_2$ mixtures show a qualitatively similar behaviour, though the blowoff branch is significantly steeper than in the case of methane-air mixtures. As well as for methane-air mixtures, stable flames were observed only for equivalence ratios noticeably exceeding the theoretical limit for the planar flame (Table 1).

A further increase of the hydrogen content in the fuel gas, up to 25%, qualitatively alters the flame blowoff behaviour. As seen from Fig. 2, the blow off is observed only within a narrow range of equivalence ratios. The blow off velocity is not a single-valued function of the equivalence ratio for these flames: a narrow range of equivalence ratios exists, in which triple blowoff limits by flow velocity, are observed: stable flames may exist only between lower and intermediate limit velocities, and above the upper limit. Apparently, a fourth blowoff limit should also exist at a velocity value above the maximum velocity achievable in the present experiments.

Experimental stabilisation limits for $0.4H_2$, $0.6H_2$, $0.8H_2$ fuel mixtures and for hydrogen-air flames are shown on the Fig. 3. Three characteristic segments on the stabilisation curves obtained for these mixtures are marked as "**a, b, c**" for the case of $0.8H_2$ flames (segment "**c**" is not present on the curve for $0.4H_2$ mixtures). Branches "**a**" correspond to the flame penetration limit. Branches "**c**" correspond to the normal blowoff limit, when flame blowoff occurs at increased flow velocity. The flame behaviour corresponding to braches "**b**" on experimental stabilisation curves is unusual: stable flames are observed only when the velocity exceeds some critical value, while at flow velocities below than this value flames cannot be stabilised. Thus, blowoff in this case occurs when flow velocity is eventually reduced, which is opposite to the normal blowoff behaviour.

As seen from Fig. 3 and Table 1, flame stabilisation in $0.4H_2$ mixtures was only possible at equivalence ratios above the theoretical LFL for the planar flame. For $0.6H_2$ mixtures, almost the entire blowoff branch "**b**" corresponds to equivalence ratios below the theoretical LFL for the planar flame. In hydrogen-air mixtures, the flame penetrated the tube at equivalence ratio below the theoretical LFL for the planar flame. Therefore, all obtained experimental points are below this limit.

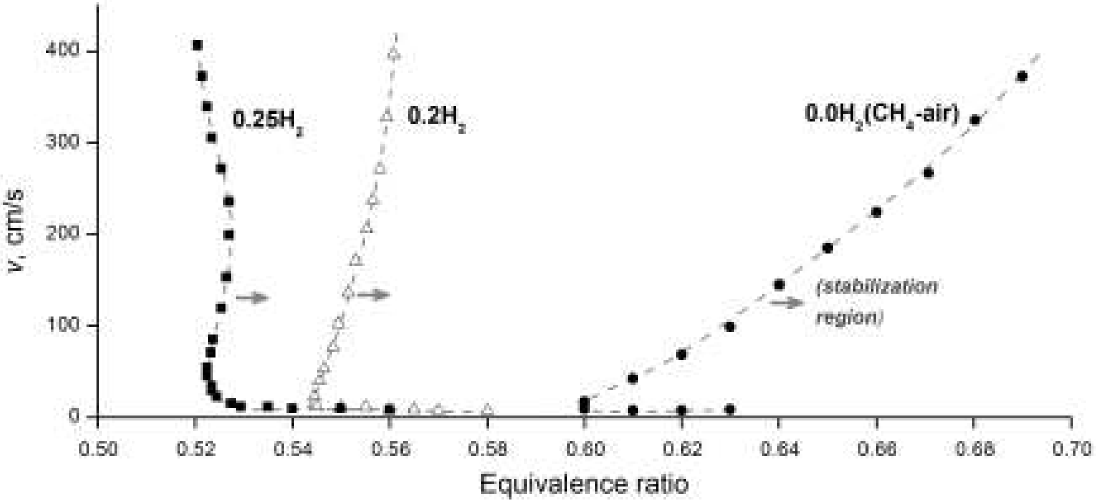


Figure 2. Flame stabilisation limits for methane-air, $(20\%H_2 + 80\%CH_4)$-air, and $(25\%H_2 + 75\%CH_4)$-air mixtures.

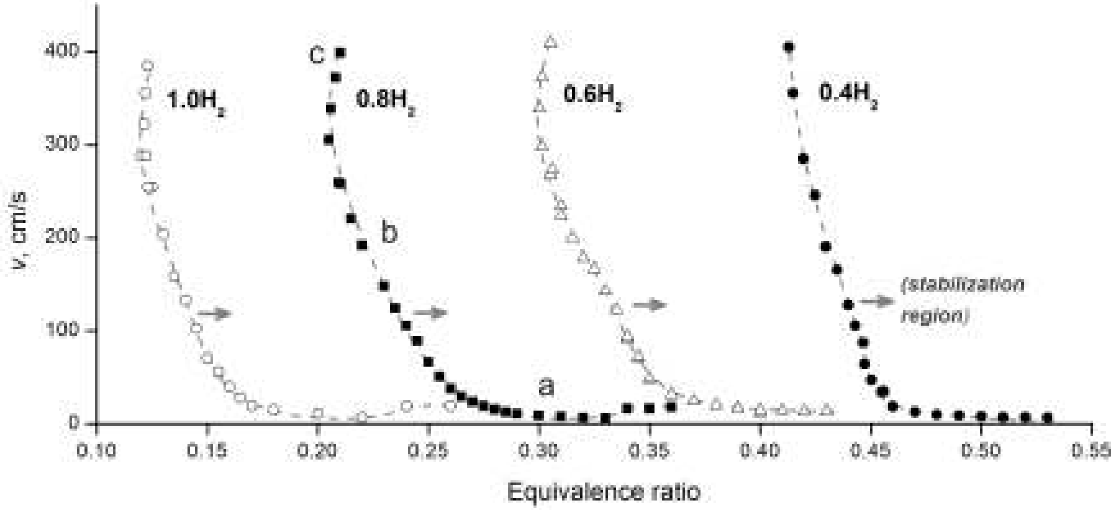


Figure 3. Flame stabilisation limits for $(40\%H_2 + 60\%CH_4)$-air, $(60\%H_2 + 40\%CH_4)$-air, $(20\%H_2 + 80\%CH_4)$-air, and hydrogen-air mixtures.

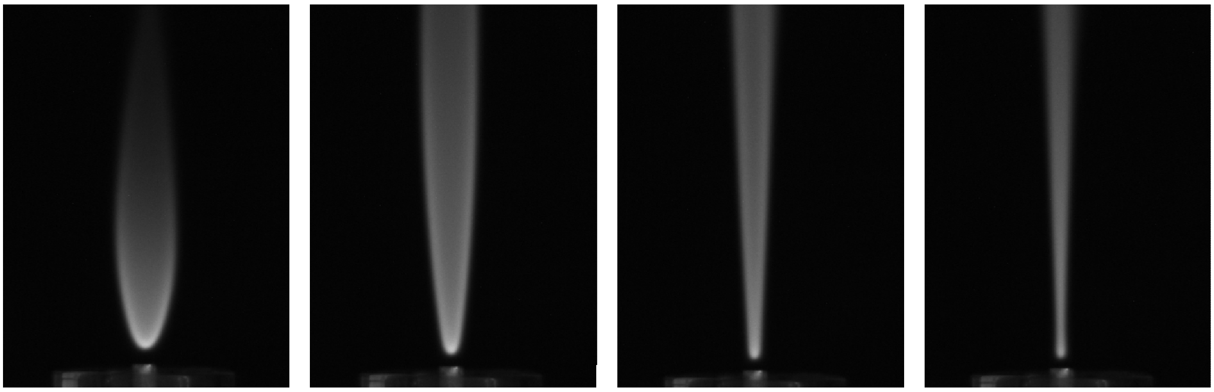
Table 1. Theoretical lean flammability limits for planar 1-D flame in methane-air, hydrogen-methane-air [10], and hydrogen-air mixtures [11].

Fuel gas	CH4	$0.2H_2 + 0.8CH_4$	$0.4H_2 + 0.6H_4$	$0.6H_2 + 0.4CH_4$	H2
LFL, planar flame, φ	**0.49**	**0.44**	**0.40**	**0.36**	**0.3**

For all tested mixtures, the measured temperature of the rod end segment did not exceed 10K near the flame blowoff limit, suggesting that heat losses to the rod did not play significant role in the flame stabilisation at near blowoff conditions. It was experimentally established earlier that heat transfer plays a negligible role in the stabilisation of inverted flames in methane-air and propane-air mixtures [4, 5, 6]. This observation was the basis for drawing the conclusion about the leading role of flame stretch effects in the inverted flames stabilisation in methane-air and propane-air mixtures [5, 6]. Thus, flames studied in the present work are also likely to be stabilised due to flame stretch effects, at least near the blowoff limits

The anomalous blowoff behaviour, found for $\geq 0.4H_2$ flames is, most likely, related to the small values of their effective Lewis numbers, and, therefore, strong, combined flame stretch/preferential diffusion effects occur, probably leading to a strong increase of the flame temperature with increasing flame stretch rate. According to results of [8, 12], the burning velocity is also expected to increase with increasing flame stretch rate in lean $0.6H_2$, $0.8H_2$, and hydrogen-air mixtures, and to be nearly insensitive to the flame stretch rate in lean $0.4H_2$ mixtures. Higher stretch rates and, therefore, higher flow velocities are needed to stabilise learner flames in such mixtures, assuming that stretch rate near the flame base increases with increasing flow velocity. Due to the positive stretching, flames far below the theoretical flammability limit for the planar flame can be stabilised in mixtures with 60% and more hydrogen in the fuel blend.

It is seen from Fig. 3 that minimal values for the equivalence ratio are observed for $0.6H_2$, $0.8H_2$ and hydrogen-air flames A hypothetical reason for the existence of these absolute limits (word “absolute” is used here in a narrow sense, as related to the particular experimental configuration used in this work) may be judged by observing the evolution of near-limit flame shapes, when these limits are approached at decreasing equivalence ratios along the branch “**b**”. Figure 4 illustrates such an evolution for $0.8H_2$ flames.



$\varphi = 0.26$
$v = 24$ cm/s

$\varphi = 0.24$
$v = 103$ cm/s

$\varphi = 0.22$
$v = 190$ cm/s

$\varphi = 0.21$
$v = 274$ cm/s

Figure 4. Near-limit flames in $0.8H_2$ mixtures at different equivalence ratios. Corresponding equivalence ratios and flow velocities are shown below each photograph. The last photograph corresponds to the absolute stabilisation limit for $0.8H_2$ flames observed in this work.

As seen from Fig. 4, near-limit flames at equivalence ratios above the absolute flammability limit have a distinguishable flame fronts at both sides of the symmetry axis. When the absolute flammability limit is nearly reached, the fronts merge, forming a narrow neck above the flame base. All flames corresponding to the normal blowoff limit (branches "**c**" on Fig. 3) looked similar to the left flame in Fig. 4. Except for these branches, fronts have never been observed to merge in the present experiments. The observed front merging closely resembles the extinction behaviour of Le < 1 counterflow flames. In coumterflow Le < 1 flames, increasing of the stretch rate at a fixed composition or decreasing equivalence ratio at a fixed flow speed, causes two opposite flat fronts to merge and leads to extinction due to incomplete reaction [13]. Similarly, front merging in near-blowoff flames in the present experiments occurs at decreasing equivalence ratio and increasing flow velocities. Thus, judging from this similarity, the likely reason for the existence of the absolute flammability limits, and for normal flames blowoff (branch "**c**") at high hydrogen content in the fuel gas is incomplete reaction in the merged fronts.

According to the suggested mechanism, the blowoff of the flames corresponding to the normal blowoff branch "**c**" on Fig. 3 should occur via breaking the neck above the flame base. There are experimental evidences that this, indeed, happens. When a normal bowoff limit, corresponding to the branch "**c**" for $0.6H_2$, $0.8H_2$ and $1.0H_2$ flames, is approached at eventually reduced equivalence ratio, erratic flame extinction and re-ignition occurs very near the blowoff limit, suggesting that a little hot spot still survives at the base of the flame. In the hydrogen-air flame, at near-maximum tested flow velocities, a very small flame above the rod edge survives even after complete extinction of the "main" flame. Figure 5 shows a photograph of such a flame. This small flame exists only within a very narrow interval of equivalence ratios below the blowoff limit ($\Delta\varphi \sim 0.001$).

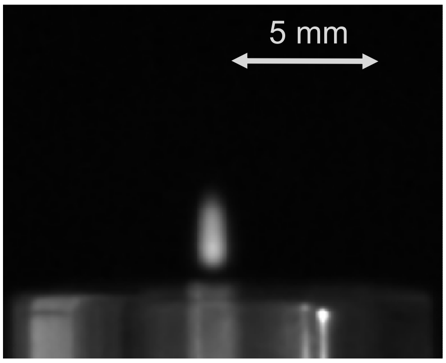


Figure 5. Small residual H_2-air flame stabilised above at the edge of the rod at the equivalence ratio just below normal blowoff limit. ($\varphi = 0.122$, $v = 384$ cm/s.)

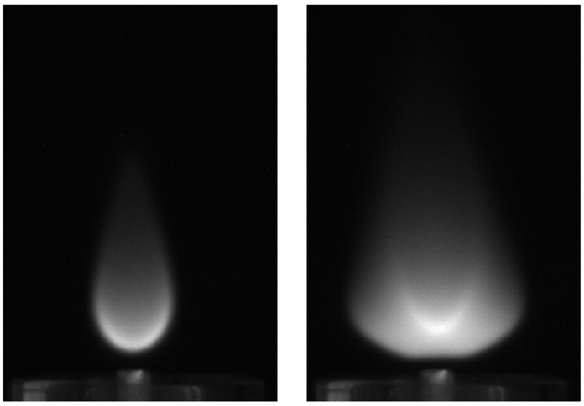


Figure 6. Stable and unstable flames in $0.8H_2$ mixtures. Left: $\varphi = 0.32$, $v = 10$ cm/s. Right: $\varphi = 0.3$, $v = 18$ cm/s.

Some comments should be made about the low-speed branch "**a**" of stabilisation limits curves shown on Fig. 4. It is seen from Fig. 4, that the minimum velocity required to stabilise H_2 -air and $0.8H_2$ flames begins to increase with increasing equivalence ratio when the right end of the branch "**a**" is approached. This is a result of the loss of stability by the corresponding flames at low flow speeds. The flame becomes spinning, or, possibly, oscillating with a frequency too high to be resolved with the available video camera or by a visual observation. Figure 6 shows typical images of stable and unstable flames corresponding to the branch "**a**" for $0.8H_2$ mixtures. The image of the unstable flame is time-averaged.

Conclusions

Inverted flames in hydrogen-methane-air flames at high hydrogen content in the fuel blend can be stabilised far below the theoretical lean flammability limits for the 1-D planar flame.

Ultra-lean hydrogen-methane-air inverted flames with hydrogen content of 40% and more in the fuel gas were found to exhibit anomalous blow-off behaviour at moderate mixture flow velocities. The blowoff of these flames occurs at decreased flow velocity, after some critical value of the velocity is achieved. Below this value, a stable flame does not exist, and above this value the flame can be reliably stabilised in a wide range of flow velocities.

Low value of measured increments of the stabilisation rod temperature above room temperature suggest that these flames are stabilised due to flame stretch effects rather than by heat losses to the rod.

The observed behaviour of inverted hydrogen-methane-air flames at high hydrogen content in the fuel blend can be attributed to strong combined flame stretch/preferential diffusion effects. Due to these effects, flame temperature strongly increases with the increase of the flame stretch, when the mixture flow velocity is increased. At the same time burning velocity in such mixtures may also increase with increasing stretch rate. As a result, learner flames are stabilised at higher flow velocities. To our knowledge, theoretical works able to explain the observed anomalous flame blowoff for $L < 0$ flames have not been published. Therefore, new theoretical investigations are necessary to explain the observed phenomena.

Normal blowoff limits for flames with high hydrogen content in the fuel blend (60% and more) were also observed in the experiments near lowest equivalence ratios for which flame still could be stabilised. Based on the observed flame shape evolution when approaching these limits, it is suggested that normal blowoff of these flames occurs due to flame fronts merging at an increased flame stretch rate. According to the suggested mechanism, merging of the flame fronts results in the incomplete reaction and flame extinction, as it takes place in Le < 1 couterflow flames at increased flames stretch rates.

Acknowledgement

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