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Experimental and numerical study of cap-like lean limit flames in H_2 - CH_4 -air mixtures

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ABSTRACT

Lean limit flames of H_2 - CH_4 -air mixtures stabilized inside a tube with an inner diameter of 30 mm in a downward flow are studied experimentally and numerically. A transition from bubble-like flames, with a long decaying skirt, to cap-like flames with a sharp visible flame edge at the bottom is observed as the lean flammability limit is approached. This transition is accompanied by formation of a secondary weak flame front inside the cap-like flame. The CH^* chemiluminescence distribution of the studied flames is recorded and the velocity field of the lean limit flames is measured using Particle Image Velocimetry (PIV). The flame temperature field is measured utilizing the Rayleigh scattering method. Numerical prediction with a mixture-averaged transport model and skeletal mechanism for CH_4 qualitatively reproduces the above experimentally observed phenomena. The presence of negative flame displacement speed for the entire leading edge of the cap-like flames is numerically predicted and experimentally demonstrated. The secondary weak flame front is located in a region with reverse upward flow of the recirculation zone, which is found to support the propagation of the leading edge with a negative flame displacement speed. Furthermore, radiative heat loss has a significant influence on the lean flammability limit of the cap-like flames.

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1. Introduction

Understanding of combustion behaviour near the lean flammability limit is of interest from both fundamental and practical points of view. The combustion behavior near the lean combustion limit are complex and affected by the flow and boundary conditions. One of the conventional combustion limit test systems is the so-called standard flammability tube introduced by Coward and Jones [1] with the following geometrical specifications: an inner diameter of 51 mm, 1800 mm length, closed upper end and open bottom end. The combustion limits in this device are determined as the lowest or highest fuel concentration in a mixture at which the flame still can propagate to the upper end after the ignition at the bottom side.

For near-unity Lewis number mixtures, the upward propagating lean limit flame looks similar to a low-density bubble rising in a tube filled with a high-density liquid, showing a spherical cap followed by a long skirt [2,3], as shown in Fig. 1. The propagation

speed of the lean limit methane/air flame is determined by buoyancy [2] and is close to that of a rising bubble which can be estimated by an analytical formula [4]. Buckmaster and Mikolaitis [5] introduced a flammability limit model for upward propagation lean methane/air flames in a standard flammability tube which was based on the assumption that flame stretch renders flame propagation impossible when it reduces the local flame speed at the flame leading point to zero. It should be noted, however, that the authors admitted the possibility of negative flame displacement speed in the case of highly stretched flames burning between counter-flowing streams of a fuel-lean mixture and that of hot combustion products, while they considered such situation in the case of the leading edge of the lean limit flames as very unlikely. According to these authors, the rising flames would be extinguished at the tip by the reverse flow of combustion products, which would be cooled by the tube wall when the recirculating gas passes by it. Later experimental measurements and numerical simulations [6] showed that the local flame speed at the leading point of the lean limit methane/air flame in a standard flammability tube is indeed close to zero. The effect of tube diameter on the extinction limit of premixed flames was experimentally studied by Babkin et al. [7], showing that the rich extinction limit for

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Fig. 1. Upward propagating bubble-like (left) and cap-like (right) lean limit flames in the 35 mm diameter tube taken from Ref. [13].

propane/air flames depends on the tube diameter. Von Lavante and Strehlow [6] investigated the local flame stretch rate distributions for lean limit methane/air flames in a vertical tube by a semi-empirical numerical model, and found that the maximum stretch rate occurs at the flame top for the upward propagating limit flames in methane/air mixture. Shoshin et al. [8,9] experimentally measured the gas velocity field for lean limit methane/air flames in the standard flammability tube using PIV, confirming that the maximum stretch rate occurs at the flame tip. However, a different mechanism of the lean limit methane/air flame was suggested by Shoshin et al. [8,9], who pointed out that local radiative heat loss becomes significant due to the formation of stagnation zone of combustion products near the flame tip, which leads to a local low temperature. Higuera and Muntean [10,11] numerically investigated the effects of tube diameter and radiative heat loss on the lean limit methane/air flame in a vertical tube using a thermal-diffusive model, and confirmed the radiative extinction hypothesis. Shoshin et al. [12] studied lean limit methane/air flames propagating in a 24 mm diameter tube and found that the lean limit is lower than that for the standard flammability tube and that the local flame speed is negative around the leading point of the flame front. The flame front has a bubble-like shape with a spherical cap and a long decaying skirt, with a relatively small recirculation zone located inside the flame near its leading point. Because the recirculation zone is separated from the tube wall by the flame skirt, the counterflowing products are not cooled significantly to cause flame extinction at the flame tip.

All the above studies focus on methane/air and propane/air flames. However, the extinction mechanism and near-limit combustion behavior in vertical tubes for hydrogen blended fuels are more complex, in particular due to the strong coupling effects of flame stretch and preferential diffusion. Shoshin and de Goeij [13] examined the tube diameter effects on the lean flammability limit for methane/hydrogen/air flames, observing different lean limit combustion regimes. Specifically, along with long bubble-like flames, with shapes similar to these of limit methane-air flames in flammability tubes, the authors also observed short

cap-like lean limit flames with a sharp bottom edge, and enclosed ball-like lean limit flames. While ball-like flames were formed in hydrogen-containing fuel mixtures in small (9–12 mm) diameter tubes, cap-like flames were observed for near lean limit mixtures in 25–50 mm diameter tubes, for a hydrogen content in the fuel gas from 10 to 40% (the maximum content used in those experiments), as shown in Fig. 1. The observed steadily propagating cap-like flames had diameters of about half the tube diameter and were symmetric relative to the tube centerline. Thus, cap-like flames are observed at the lean flammability limit for mixtures with a wide range of Lewis number, $Le < 1$, and this fact makes studying this combustion regime important. Besides, the combustion mechanism of such flames is relevant to propagation of gravity-affected near-limit low Lewis number flames [14] in conditions different from the ones studied here. An interesting observation made in [13] was the presence of a secondary concave cone-like shaped front inside the main front of the cap-like flames. The presence of this front and its possible role in the cap-like flame propagation remain, however, unexplained, and the combustion regime of the observed cap-like flames is not fully understood.

The main objective of the present study is to investigate the structure and combustion mechanism of the cap-like lean limit flames propagating upward in a tube. In our experiments, hydrogen/methane/air flames are stabilized in a downward flow with an inlet velocity kept near the flame blow-off limit which closely corresponds to the upward flame propagation speed in a tube of the corresponding diameter. While a stationary flame burns in nearly the same conditions as the one propagating upward in the tube,¹ it allows for more detailed experimental measurements. A tube with an inner diameter of 30 mm has been selected for the experiments. The 30 mm diameter tube is chosen because it was found that, for this tube diameter, the cap-like flames can form for a wide range of hydrogen fractions in methane (wide range of Le) [13]. Another objective of this work is to study the Lewis number effect on the cap-like lean limit flames. For that purpose, hydrogen-methane fuel gas mixtures, with hydrogen contents of 20 and 40% by volume, are used in the experiments and the simulations. The Abel inverted CH^* chemiluminescence of the studied flames is recorded as a function of equivalence ratio, ϕ . The velocity field inside the tube is measured experimentally using the Particle Image Velocimetry method (PIV). The flame temperature fields are experimentally determined by the Rayleigh scattering method. Detailed numerical simulations of the cap-like lean limit flames are performed with a mixture-averaged transport model and skeletal chemical mechanism for methane of Smooke [15]. In the following sections, the experimental setups and numerical modeling are described. Subsequently, the experimental and numerical results are discussed.

2. PIV measurements

A schematic of the experimental setup for PIV measurements is shown in Fig. 2. A quartz tube with an inner diameter of 30 mm, length of 50 mm and wall thickness of 1.5 mm is connected to a plenum chamber. The flow is preconditioned by the metal foam plate installed inside the plenum chamber. A 30 mm diameter perforated plate with thickness of 2 mm and holes of 0.5 mm

¹ It should be noted that the burner-stabilized cap-like lean limit flames are affected by the boundary layer. However, the thickness of this boundary layer in the entire domain is relatively small, because of the speed-up of the hot gas near the tube wall. The slightly larger thickness of boundary layer does not introduce qualitative changes in the structure of the burner-stabilized cap-like flame, as compared to the cap-like flame propagating in a tube. Nevertheless, it does introduce some quantitative differences. The presence of the thin boundary layer has a similar effect as the decrease of tube diameter. Therefore, it is reasonable to observe that the blow-off limit velocity is lower than the rising bubble speed in the same diameter tube [13], as the rising speed decreases with the decrease of tube diameter.

Fig. 2. Schematic of experimental PIV setup.

diameter uniformly spaced with 0.3 mm pitch, separates the chamber and the tube and produces a uniform mixture flow at the tube inlet side. A flame can be stabilized inside the quartz tube after ignition below the tube.

In order to eliminate the background reflection from the laser light, the back part of the inner tube wall is painted with thermal resistant black paint. For the front part of the tube wall, two small parts of outer tube wall near the laser sheet are also covered with the black paint. The mixture is seeded with Al_2O_3 particles with an average size of approximately $1\ \mu\text{m}$ using a custom-made fluidized bed seeding system. The flame region is illuminated by a light sheet of an Nd:YAG pulsed laser with a wavelength of 532 nm. As reported in the following section, the flame displacement speed at the leading edge of the limit flames is negative, meaning that a stagnation surface is located at the upward side of the reaction zone. Due to this, streamlines which originate from the fresh mixture, as well as seeding particles carried by the mixture, do not penetrate the flame front. All the particles are transported out of the tube by the flow near the tube wall. In order to introduce particles into the flame with the reverse upward flow in a recirculating zone, a thin metal strip connected with an oscillating motor is installed at the outlet of the tube. The slowly oscillating metal strip slightly disturbs the flow at the tube outlet and introduces particles from the periphery to the middle part of the tube. The particles are then transported towards the flame region due to the flame generated recirculation zone, as shown in Fig. 3. Even with this remedy, the amount of seeding particles in the flame is not sufficient to recover the velocity field from a single measurement. In order to measure the entire flow field, multiple pairs of consecutive PIV images are stacked into one pair and then processed. The effects of the oscillating metal strip on the flow field in the flame region are almost negligible. This statement is based on the observation that the introduction of the strip has no observable influence on the flame shape, size and position. The post-processing of raw PIV images is performed using the PIVview2C software [16]. The fuel compositions and inlet velocities considered in the experiments for PIV measurements and for Rayleigh temperature measurements are listed in Table 1. The inlet velocities in the experiments for different fuel compositions are chosen near the blow

Fig. 3. A pair of consecutive PIV images of the flame in the (40% H_2 + 60% CH_4)-air mixture at an equivalence ratio of 0.31.

Table 1

Summary of conditions for the experiments with different equivalence ratios. Effective fuel Lewis number (Le) for CH_4 and H_2 mixtures is evaluated by taking a mole average.

Condition	Fuel	Le	Inlet velocity (cm/s)
I	20% H_2 + 80% CH_4	0.84	11.8
II	40% H_2 + 60% CH_4	0.7	13.7

off limit of the lean limit flames (determined in separate experiments), which are very close to the upward flame propagation speed in a tube of corresponding diameter.

3. Rayleigh temperature measurements

A schematic of the experimental setup for Rayleigh scattering temperature measurements is shown in Fig. 4. The temperature field of the entire flame is measured by line-wise Rayleigh scattering using a burner that can traverse a steadily burning flame inside

Fig. 4. Schematic of experimental setup for Rayleigh temperature measurement.

the tube through the measurement section. This burner consists of a hollow tubular piston and a quartz tube with an inner diameter of 30 mm and a length of 220 mm. A linear motor moves the piston inside the tube with a constant speed. A perforated plate with thickness of 2 mm and holes of 0.4 mm diameter uniformly spaced with 0.5 mm pitch is mounted in the outlet of the piston to generate a uniform outlet flow profile. The flow is pre-conditioned by a metal foam disc installed above the perforated plate inside the piston. The mixture is supplied through two side inlets at the upper part of the piston. The slit between the piston and the tube is sealed by a circular rubber seal, which tightly envelops the tube and the piston.

A CW diode laser with an output power of 1 W operated at 450 nm is used for the measurements. In order to avoid strong background scattered light when the laser beam passes through the tube wall, two symmetrical holes with a diameter of 2 mm are drilled at the position of 100 mm from the top end of the quartz tube. Two metal tubes with an inner diameter of 6 mm are horizontally attached to the tube wall sharing the co-axis with both small holes. At the inlet side of the laser beam, the metal tube is fitted with an anti-reflection window, while a Brewster window is mounted at the outlet side of the laser beam. The inner metal tube wall and half of the inner quartz tube wall are painted with black heat resistant paint. The laser beam is focused in the center of the tube with a 20 mm focal length lens, procuring a nearly constant width beam of 0.5 mm diameter inside the quartz tube. A flame inside the tube is scanned by moving the piston with a constant speed of 0.7 mm/s. The scattered light from the laser beam inside the tube is recorded by a AVT-PIKE F-032b CCD-camera with a frame rate of 8 fps. The camera lens is equipped with an interference filter (center wavelength 450 nm and bandwidth 10 nm) to filter out flame radiation. The Rayleigh scattering images recorded during the piston motion are utilized to reconstruct the temperature field inside the tube using the known image scale and frame rate, achieving a resolution of 0.05 mm in horizontal direction and 0.175 mm in vertical direction.

The details on the Rayleigh scattering experiments and data processing can be found in our previous publication [17]. The effective Rayleigh scattering cross section for the present lean limit

flames is evaluated using the numerically predicted distribution of species in the studied flames, as shown in Fig. 5. The effective Rayleigh scattering cross section is scaled by the effective Rayleigh cross section for the corresponding unburned mixture, as the calibration of the measurements is performed using the corresponding unburned mixture. Figure 5 shows that the influence of variations of the effective Rayleigh scattering cross section over the flame region on the precision of temperature measurements is small. Averaged effective cross-section between its minimum and maximum values in the simulated flames is taken. The estimated error, based on Fig. 5, indicating by the variations of the effective cross-section does not exceed 1% and 0.5% for (20% H₂+80% CH₄)-air flames and (40% H₂+60% CH₄)-air flames, respectively.

Flame chemiluminescence is recorded at a fixed piston position at a distance of 50 mm from the outlet of the tube. An AVT-PIKE F-032b CCD-camera equipped with an interference filter (430 nm and bandwidth 10 nm) is employed to record the CH* chemiluminescence radiation. The chemiluminescence images show a line-of-sight integrated emission intensity. The in-plane radial emission intensity distributions are recovered by Abel inversion of the horizontal recorded intensity profiles.

For both PIV and Rayleigh temperature measurements, cylinder gases (H₂, CH₄, synthetic air) with 99.9% purity are used in the experiments. Mass flow controllers with an uncertainty less than 1% are employed to control the supply of H₂, CH₄ and synthetic air. Small fluctuations of mixture compositions induced by inherent instability of the mass flow controllers are damped by a buffer mounted in the gas line before the mixture is supplied to the burner. The mixture is ignited at the bottom of the burner at a sufficiently large equivalence ratio using a household lighter. A lean limit flame can be stabilized inside the tube by decreasing the equivalence ratio. The tube wall is kept nearly at room temperature (< 325 K) by supplying cooling air to the outside of the quartz tube. The tube wall temperature is monitored by a pyrometer Pyrospot DT 40L.

4. Numerical approach

The laminar and axisymmetric flames under study are mathematically described by the conservation equations of mass, momentum, energy and species in a cylindrical coordinate system, as described in our previous publication [18]. The flow is treated as a mixture of perfect gases under the assumption of an ideal gas equation of state. The set of governing equations is solved by the body-fitted, multi-block, adaptive mesh refinement, finite-volume framework that has been originally developed by Groth and co-workers [18–20]. Thermodynamic properties, transport properties and species net production/destruction rate are all computed by the open-source library CANTERA [21].

In the simulations, the computational domain is a cylindrical channel with a radius of 15 mm and a height of 50 mm, as shown in Fig. 6. The left boundary of the domain aligns with the axis of symmetry. The upper boundary is assumed to be isothermal with a temperature of 300 K. The upper boundary also accounts for the diffusion of fuel/air mixture across the boundary. The fuel/air mixture is supplied uniformly from the upper boundary. The right boundary is treated as a non-slip and iso-thermal boundary with a temperature of 300 K according to measured temperatures of the tube wall. The lower boundary is modelled as a free outflow. The diffusion velocity is computed using the mixture-averaged transport model. The skeletal mechanism for CH₄ of Smooke [15] is employed in the present simulations, which consists of 16 species and 25 elementary reactions. These chemical and transport models yield good qualitative results in the simulation of ball-like lean limit flames in our previous research [22]. Gravity and radiative heat loss are taken into consideration. Thermal radiation is

Fig. 5. Scaled effective Rayleigh scattering cross section for the lean limit flames. a. 20% H₂; b. 40% H₂. The effective Rayleigh scattering cross section for lean limit flames is scaled by the effective Rayleigh scattering cross section of the unburned mixture at 1 bar and 300 K.

Fig. 6. Schematic of the computational domain and boundary conditions.

modelled via the optically-thin Planck model. The Planck mean absorption coefficient of the mixture is evaluated from the major radiating species CO₂, H₂O and CO. The individual coefficients are calculated using a statistical narrow-band model and the dataset of Soufiani and Taine [23]. The solutions are obtained with up to three levels of mesh refinement, achieving a minimum resolution of 54 μm for all the cases. These levels of resolution are found to be sufficient to get grid-independent solutions due to the relatively large flame thickness for flames near the lean limit. More details of the numerical method can be found in [18]. The conditions of the simulated cases are listed in Table 2.

5. Results and discussion

5.1. Flame shape and overall structure

Figure 7 displays the Abel inverted CH* chemiluminescence distributions for the flames with different H₂ contents in the fuel gases as a function of equivalence ratio (ϕ). For both H₂-containing mixtures, a transition from bubble-like flames, with a long decaying skirt (open trailing edge), to cap-like flames with a sharp vis-

Table 2

Summary of conditions for the simulated cases with the tube diameter of 30 mm.

Case	Fuel	V_{in} (cm/s)	ϕ
A	20% H ₂ + 80% CH ₄	11.8	0.440
B	20% H ₂ + 80% CH ₄	11.8	0.410
C	20% H ₂ + 80% CH ₄	11.8	0.395
D	20% H ₂ + 80% CH ₄	11.8	0.390
E	40% H ₂ + 60% CH ₄	13.7	0.400
F	40% H ₂ + 60% CH ₄	13.7	0.340
G	40% H ₂ + 60% CH ₄	13.7	0.330
H	40% H ₂ + 60% CH ₄	13.7	0.315

ible flame edge at the bottom (closed trailing edge) is observed with the decrease of equivalence ratio. This transition is accompanied by formation of a secondary, weak flame front inside the cap-like flame. The secondary flame front connects the bottom edge of the main front of the flame, and, as well as the main one, is convex toward the burner inlet side. It is seen in Fig. 7 that the CH* luminosity of the secondary flame front for the (40% H₂+60% CH₄)-air flame is weaker than that for the (20% H₂+80% CH₄)-air flame, indicating a lower burning rate of CH₄. Also, the size of the lean limit flame in the (20% H₂+80% CH₄)-air mixture is smaller than that in the (40% H₂+60% CH₄)-air mixture. A weak visible line which is convex toward the downstream side in some flames is caused by the Abel inversion error, because of slight asymmetry of the bottom edge of the flames.

The lean flammability limits obtained in the experiments for the mixtures with 20% and 40% H₂ contents are $\phi = 0.395$ and 0.31, respectively, which are in agreement with those measured in long vertical tubes with inner diameter of 25 and 35 mm [13]. A slight decrease of the equivalence ratio below the lean limit value leads to flame extinction. During the quenching process, the bottom edge of the flame collapses toward the middle of the flame, forming, at the final stage, a ball-like flame, which propagates upward and then quenches when it touches the perforated plate.

Figure 8 displays the measured velocity field for the lean limit flames in the mixtures with 20 and 40% H₂ contents. The flame front in Fig. 8 is determined by the maximum luminosity of the flame image which is visible on the PIV images (Fig. 3). The lean limit flame containing 20% H₂ ($\phi = 0.395$) is extinguished by the PIV particles in the experiments. Therefore, the PIV measurement

Fig. 7. The Abel inverted CH^* chemiluminescence distributions for different equivalence ratios for (20% H_2 +80% CH_4)-air flames (Upper) and (40% H_2 +60% CH_4)-air flames (Lower).

Fig. 8. Experimentally measured velocity fields for the lean limit flames with different H_2 contents. a. (20% H_2 +80% CH_4)-air flame; b. (40% H_2 +60% CH_4)-air flame. The black line denotes the main flame front.

Fig. 9. Distribution of chemical heat release (W/m^3) and flow streamlines for the flames studied in the simulations with different H_2 contents in the fuel gases. Upper: (20% H_2 +80% CH_4)-air flames; Lower: (40% H_2 +60% CH_4)-air flames.

for the lean limit flame of (20% H_2 +80% CH_4)-air mixture is performed at $\phi = 0.397$, which is found to be the lean limit for the seeded mixture. It can be seen in Fig. 8 that the entire lean limit flame in both H_2 containing mixtures is located inside a recirculation zone, reflecting that no fresh mixture is delivered to the reaction zone by convection across the stagnation surface. In the main flame front, the transport of the unburned mixture below the stagnation surface to the reaction zone is solely caused by diffusion against the convective flow of combustion products. The local gas velocity in the reaction zone is directed against the velocity of the fresh mixture flow, which is characterized as a negative flame displacement speed [24]. The local flame speed of the secondary flame front can be characterized as positive if its propagation relative to the gas flowing upward in the recirculation zone is considered. A negative flame displacement speed was first experimentally observed in counterflow premixed flames [25,26]. Also, a possibility for the occurrence of a negative flame displacement speed in a lean limit CH_4 -air flame propagating in a standard flammability tube is theoretically predicted in Ref. [5]. However, the authors claimed that the steady state upward propagation lean limit flame with negative flame displacement speed is not experimentally observable, because, according to the authors, the flame would be extinguished by the flux of cooled combustion products into the reaction zone. The speculations of the authors, however, were related to CH_4 -air flames, for which the Lewis number is close to unity and flame-stretch-induced preferential diffusion effects are small. In low Lewis number mixtures, however, the enhancement of the local flame burning intensity at the flame leading edge due to strong preferential diffusion can, at least to some extent, compensate the cooling effect of the counterflowing combustion products. An additional crucial factor that makes survival of the

cap-like flames with negative flame speed possible is the presence of the secondary flame front inside the cap-like flames, which supports the leading front by providing an additional amount of flame heat. The secondary flame front is formed, because a large fraction of the unburned mixture leaks from the upstream side to the downstream side. This will be clarified by numerical results in the following paragraphs.

Figure 9 displays the simulated distributions of chemical heat release rate and streamlines for the flames with different H_2 contents at different equivalence ratios. It is seen that the experimentally observed transition from bubble-like flame to cap-like flame is qualitatively predicted by the numerical simulations. Both in experiments and simulations, this transition happens suddenly when the inlet equivalence ratio is reduced, while no intermediate regime is observed. It also can be seen in Fig. 9 that a recirculation zone occurs in the flames as the lean limit is approached, which is due to the fact that buoyancy-induced natural convection becomes important near the lean limit as the flame burning velocity becomes smaller. Consistent with the experimental results, the simulated lean limit flames for both H_2 -containing mixtures are entirely located inside a recirculation zone. Besides, the occurrence of a recirculation zone is accompanied by the appearance of a local minimum of the heat release rate at the flame tip for the (20% H_2 +80% CH_4)-air flames, while this phenomenon is not observed for the (40% H_2 +60% CH_4)-air flames. This phenomenon can be explained by the coupling effect of radiative heat loss and preferential diffusion. For both cap-like lean limit flames, the tangential speed along the flame front has the lowest values near the flame tip. Consequently, the gas near the flame tip is effectively cooled by radiative heat loss. For the (20% H_2 +80% CH_4)-air flames, the reduction in burning intensity at the flame tip caused

Fig. 10. Mass fraction distributions of CH_4 , H_2 and O_2 for the flame with 40% H_2 content at $\phi = 0.315$.

by radiative heat loss dominates the increase in burning intensity at the flame tip caused by the coupling effects of positive flame stretch and preferential diffusion, thus a local minimum appears in the heat release rate at the flame tip. Conversely, for the (40% H_2 +60% CH_4)-air flames, with more H_2 content in the fuel gases, the opposite situation takes place and the heat release rate peaks at the flame tip. The validity of this argumentation will be further demonstrated in Section 5.5 by performing detailed numerical simulations without radiative heat loss.

The predicted lean flammability limit in the mixture with 20% H_2 content is obtained at $\phi = 0.39$, which is slightly lower than the experimental value. However, the predicted lean flammability limit in the mixture with 40% H_2 content is attained at $\phi = 0.315$, which is slightly larger than the corresponding measured one. The discrepancy between experimental and numerical results is caused by the combined inaccuracies in the transport model and chemical mechanism as well as boundary conditions.

Figure 10 presents the mass fraction distributions of CH_4 , H_2 and O_2 for the lean limit flame with 40% H_2 content. It can be seen that a large fraction of CH_4 , H_2 and O_2 leaks from the upstream side to the downstream side through the gap between tube wall and flame. The species (H_2 , CH_4 , and O_2) diffuse from the peripheral near-wall flow into the recirculation zone and are then brought back to the flame region with the upward convective flow in the core of the recirculation zone. Though the unburned mixture inside the recirculation zone is highly diluted by the combustion products, the burning of this mixture is made possible by preheating: a large amount of the heat generated in the primary front is also transported to the secondary flame front by conduction.

The centerline variations of chemical heat release rate and net consumption rates of CH_4 and H_2 for the flames in two different H_2 -containing mixtures with different inlet equivalence ratios are displayed in Fig. 11. It can be seen that chemical heat release rate decreases significantly with the decrease of inlet equivalence ratio. The presence of two peaks for the heat release rate is observed for the lean limit flames. Similar to the heat release rate, both net consumption rates for CH_4 and H_2 diminish with decreasing the inlet equivalence ratio. However, only CH_4 exhibits two clear peaks for the net consumption rate, indicating that the heat release in the middle part of the secondary front of the lean limit flames is mainly caused by the consumption of CH_4 . The disappearance of H_2 at the apex of the secondary flame front can be explained by

the fact that the secondary flame front is negatively stretched, as the flow configuration there is similar to the one at the apex of a Bunsen flame. Negative flame stretch is known to weaken low Lewis number flames [27]. Because H_2 is a highly diffusive component, the preferential diffusion effect is much stronger for H_2 compared to CH_4 . This phenomenon is qualitatively similar to weakening or extinction of the tip of a Bunsen flame for low Lewis number mixtures [28].

5.2. Flame temperature

The experimentally measured (a–d) and numerically simulated (e–f) temperature fields for the flames in the mixtures with 20% (a, b, e, f) and 40% (c, d, g, h) H_2 contents at different inlet equivalence ratios are shown in Fig. 12. A qualitative agreement between measured and simulated temperature fields is observed. It also can be seen that the size of the hot core region decreases with diminishing inlet equivalence ratio for both measured and simulated flames. For the simulated flames, the maximum flame temperature for all the flames is achieved near the flame tip except for the simulated flames containing 20% H_2 in the fuel gases at the inlet equivalence ratio of 0.39. The maximum temperature along the flame centerline for both flames is about 16 K lower than the maximum flame temperature. The reason for this is radiation as has been explained in Section 5.1. The lack of a lower temperature at the flame tip in the measured temperature field can be attributed to the experimental uncertainties which exceed the 16 K.

The measured and simulated centerline distributions of the temperature for the lean limit flames with different H_2 contents are presented in Fig. 13. It is seen that the flame temperature rapidly increases at the upstream side and decreases significantly at the downstream side. Notably, the cooled low density mixture is effectively heated up when the mixture flows upward through the secondary flame front. Consequently, the temperature inside the flame maintains nearly constant. Even though the heat release rate in the secondary flame front is much smaller than in the flame leading edge, the thermal effect of the secondary flame front is significant, because the gas mass flow rate through the secondary flame front is small. The heated combustion products are then transported into the main flame front reaction zone, preventing the extinction of the main flame front. The experimental cap-like flame with 20% H_2 content is shorter than the simulated one.

Fig. 11. Profiles of chemical heat release rate (left) and consumption rates of CH_4 (middle) and H_2 (right) along the centerline for flames with different inlet equivalence ratios. Upper: (20% H_2 +80% CH_4)-air flames; Lower: (40% H_2 +60% CH_4)-air flames.

Fig. 12. Experimentally measured (a–d) and numerically simulated (e–h) temperature fields for the flames in the mixtures with 20% (a, b, e, f) and 40% (c, d, g, h) H_2 contents at different equivalence ratios.

Fig. 13. Distribution of flame temperature along the centerline for the measured and simulated lean limit flames in different H_2 containing mixtures. Upper: 20% H_2 ; Lower: 40% H_2 . Vertical dash-dotted lines denote the numerical flame front position.

Correspondingly, the measured high temperature zone is smaller than the simulated one. Also, the measured maximum flame temperature along the centerline is slightly larger than that of the simulated flame. For the cap-like flame with 40% H_2 content, the measured centerline flame temperature profile in the flame region is in good agreement with the simulated temperature, and the maximum flame temperatures match almost exactly.

5.3. Flame stretch and fuel consumption

For a stationary cylindrically symmetric flame, the local flame stretch rate, k , can be calculated using [6],

$$k = \frac{dv_t}{dL} + \frac{v_t \cos \theta}{R} \quad (1)$$

where v_t is the tangential component of the gas velocity along the flame front, L is the distance along the flame front, θ is the angle of the normal to the flame front and the flame symmetry axis, and R is the radial distance from the selected flame front location to the symmetry axis. The stretch rate can not be defined uniquely due to the large flame front thickness. From the present simulated flames, it is found that, although the value of the local flame stretch rate changes with the change of flame front location determined by different isothermal lines, the maximum local flame stretch rate always peaks at the flame tip. It is also noted that the measured local flame stretch rate distribution determined for the flame front location defined by the maximum luminosity is very noisy, because the calculation of flame stretch rate is very sensitive to v_t , and the measured v_t along this defined flame front is noisy. Consequently, the measured and simulated stretch rate distributions along the flame front are determined at the stagnation surface (isothermal line of 880 K), where the uncertainty of the measured v_t is small. The definition of the flame front at the stagnation surface was also used in Ref. [8]. Figure 14 shows the measured and the simulated local flame stretch rate distributions along the flame front for the lean limit flames. It can be seen that both measured and simulated

Fig. 14. Distribution of flame stretch rate along the flame front.

local stretch rates attain their maximum values at the top of the flames ($L = 0$). The measured flame stretch rates are slightly lower than the simulated ones, though the discrepancy is not large.

Local consumption rates for CH_4 and H_2 , $\dot{m}_\alpha^c$, are computed as:

$$\dot{m}_\alpha^c = \frac{-1}{A_c} \int_{\Omega_\alpha^{local}} \omega_\alpha d\Omega^{local} \quad (2)$$

where A_c is the length of the flame front inside the control surface, α denotes CH_4 or H_2 , ω_α is the consumption rate of species α , and Ω_α^{local} is the local control surface. The flame front for each case is determined at the iso-thermal line corresponding to the maximum heat release. The local control surface is the area between two adjacent local normals which are uniformly distributed along the flame front. The local normals follow the gradient of temperature. The local consumption rates for CH_4 and H_2 along the flame front are presented in Fig. 15 from the tip of the main flame front ($L = 0$) to the tip of the secondary flame front. It is seen that, for the flames with 20% H_2 contents, the local consumption rates of CH_4 and H_2 have local minimum values at the flame tip for both inlet equivalence ratios. This phenomenon is slightly more distinct at smaller inlet equivalence ratio, owing to the stronger radiative heat loss relative to the heat release rate with decreasing inlet equivalence ratio. However, for the flames with 40% H_2 content, the local consumption rates of CH_4 and H_2 are nearly constant over the distance from 0 to 5 mm for the flame tip, and decrease further away from the flame leading point for both inlet equivalence ratios. This can be argued as follows: with more H_2 content in the fuel gases, the increase in burning rate at the flame tip caused by preferential diffusion dominates the reduction in burning rate caused by the radiative heat loss. Figure 15 further shows that the

Fig. 15. Distribution of local flame consumption rate for CH₄ and H₂ along the flame front from the tip of the main flame front to the tip of the secondary flame front.

local consumption rates of CH₄ and H₂ at the tip of the main flame front decrease with the decrease of equivalence ratio, while those peaks at the flame bottom edge increase due to an increase in local equivalence ratio caused by flame curvature. Based on above observations, an extinction mechanism for the cap-like flames is proposed. With the decrease of equivalence ratio, the size and intensity of the buoyancy-induced recirculation zone increase, which leads to a more curved flame bottom edge. As a result, the burning rate at the bottom edge increases. Correspondingly, the burning velocity of the bottom edge increases, which causes the flame bottom edge to get closer to the flame tip and then reaches a new balance between the burning velocity of the bottom edge and the corresponding flow velocity. However, as the lean limit is reached, the balance of the burning velocity of the bottom edge and the corresponding flow velocity can not be maintained, which leads to the extinction of the cap-like flame.

5.4. Local equivalence ratio

For low Lewis number flames, the local mixture can be significantly enriched by the coupling effects of preferential diffusion and positive flame stretch/curvature, which leads to a larger local equivalence ratio. Therefore, in order to evaluate the influence of preferential diffusion on the cap-like flames, a local equivalence ratio is computed for the simulated cases based on mixture

fraction, ζ , which is given by Peters [29],

$$\phi = \frac{\zeta}{(1-\zeta)} \frac{1-\zeta_{st}}{\zeta_{st}} \quad (3)$$

where ζ_{st} denotes the stoichiometric mixture fraction. Bilger's definition [30] of mixture fraction is employed, which is expressed as:

$$\zeta = \frac{\beta - \beta_{ox}}{\beta_{fu} - \beta_{ox}} \quad (4)$$

where β is a coupling function, subscripts *fu* and *ox* denote the fuel stream and oxidizer stream, respectively. For a fuel blend of H₂ and hydrocarbon C_mH_n, with *m* and *n* referring to the number of atoms of the elements C and H, respectively, the coupling function β based on the element mass fractions is given by,

$$\beta = \frac{Z_C/M_C}{m(1-\chi)} + \frac{Z_H/M_H}{n(1-\chi)+2\chi} - \frac{Z_{O,ox}/M_O}{(1-\chi)(m+n/4)+\chi/2} \quad (5)$$

where Z_α is the element mass fraction of element α and M_α is the molar mass of α , with $\alpha = C, H$ and O , and χ is the molar fraction of H₂ in the fuel. The stoichiometric mixture fraction (ζ_{st}) is determined by the following equation:

$$\zeta_{st} = \frac{\frac{Z_{O,ox}/M_O}{(1-\chi)(m+n/4)+\chi/2}}{\frac{Z_{C,fu}/M_C}{m(1-\chi)} + \frac{Z_{H,fu}/M_H}{n(1-\chi)+2\chi} + \frac{Z_{O,ox}/M_O}{(1-\chi)(m+n/4)+\chi/2}} \quad (6)$$

Figure 16 shows the simulated distributions of local equivalence ratio scaled by the corresponding inlet value for different

Fig. 16. Local equivalence ratio scaled by the corresponding inlet value for the flames in different H_2 containing mixtures. (a, b): $(20\% \text{H}_2 + 80\% \text{CH}_4)$ -air flames and (c, d): $(40\% \text{H}_2 + 60\% \text{CH}_4)$ -air flames. The scales are different for different cases.

Fig. 17. Distribution of local equivalence ratio along the flame front from the tip of the main flame front to the tip of the secondary flame front.

H_2 contents in the fuel gases. It can be seen that the variation in equivalence ratio is more pronounced with more H_2 content and with lower inlet equivalence ratio. For the lean limit flame with 40% H_2 content, the maximum increase and decrease in equivalence ratio with respect to its inlet value are 29.5% and 3%, respectively, while these for the lean limit flame with 20% H_2

content are only 20 and 2.5%. Figure 17 displays the distribution of the local equivalence ratio for the lean limit flames with different H_2 contents in the fuel gases along the flame front from the tip of the main flame front to the tip of the secondary flame front. The flame front is defined as the iso-thermal line corresponding to the maximum heat release rate. It is seen that, for both lean limit flames, the local equivalence ratio along the flame front peaks at the main flame front tip and flame bottom edge, respectively, while it has the lowest value at the tip of the secondary flame front. The values of the local equivalence ratio along the flame front for both flames are larger than the corresponding inlet equivalence ratio, which can be explained that large fraction of unburned mixture flows downstream around the leading edge without combustion and excess H_2 from downstream diffuses back into the flame. Besides, the whole leading edge of the flame is positively stretched, so that mixture becomes enriched with H_2 in the entire flame front.

5.5. Radiative heat loss effect

To assess the effect of radiative heat loss on the local heat release rate for the lean limit flame with 20% H_2 content, numerical simulations without radiative heat losses are performed. Excluding radiative heat losses from the simulations significantly extends the flammability limit, from $\phi = 0.39$ with radiation to $\phi = 0.34$ without radiation. Figure 18 shows the simulated distribution of

Fig. 18. Distribution of chemical heat release (W/m^3) and flow streamlines for the $(20\% \text{H}_2 + 80\% \text{CH}_4)$ -air flames at $\phi = 0.39$ with and without radiative heat loss (a, b) and at $\phi = 0.34$ without radiation heat loss (c).

chemical heat release rate and flow streamlines for the lean limit flames in the (20% H₂+80% CH₄)-air mixture at $\phi = 0.39$ with radiative heat loss (a), for the flame at the same ϕ without radiative heat loss (b), and for the lean limit flame at $\phi = 0.34$ without radiative heat loss (c). It can be seen that the difference for the flame shapes simulated with and without radiative heat loss is prominent at $\phi = 0.39$. The flame shape and streamline pattern of the flame at $\phi = 0.39$ without radiative heat loss resemble those of the flames at larger equivalence ratio with radiative heat loss (Fig. 9). Shapes and sizes of the lean limit flames simulated with and without radiative heat loss (a, c) are comparable. Also, both lean limit flames are located in a recirculation zone. It should be noted that no local minimum heat release rate at the flame tip is observed for the lean limit flame without radiative heat loss. This observation demonstrates that the lower burning intensity at the flame tip for the near-limit (20% H₂+80% CH₄)-air flames (Fig. 9 (c,d)) is caused by radiative heat loss. Though the radiative heat loss affects the distribution of chemical heat release rate along the flame front as well as the value of the lean limit, it seems to be unimportant for the extinction mechanism of the present lean limit flame. Both simulated lean limit flames with and without radiative heat loss begin to extinguish from the flame bottom edge and collapse towards the leading point when the inlet equivalence ratio is set slightly below the limit value. This extinction scenario is also consistent with experimental observation.

6. Conclusions

In this work, we investigated the cap-like lean limit flames of two H₂-CH₄-air mixtures stabilized inside a 30 mm diameter tube in a downward mixture flow. As the lean flammability limit is approached, a sudden transition from bubble-like flame to cap-like flame is observed accompanied by the formation of a secondary weak flame front inside the flame. This experimentally observed phenomenon is qualitatively predicted by the numerical simulations.

Both experimental and numerical results show that the cap-like lean limit flame is located inside a recirculation zone. The entire leading edge of the cap-like flame is found to be propagating with a negative flame displacement speed. The burning of the secondary weak flame front is supported by the unburned mixture which leaks between the cap-like flame and tube wall and the released heat from the main flame front. The presence of the secondary weak flame front is a key feature of the studied cap-like flames, which makes the main flame front propagating with a negative flame displacement speed possible. The measured flame temperature is reasonably in agreement with the predicted temperature. The maximum local stretch rate is attained at the flame tip, which is similar to the CH₄-air flames in a tube. It is also found that radiative heat loss has a significant influence on the lean flammability limit of the cap-like flames. The extinction of the cap-like flame could be caused by the imbalance of the flame bottom edge burning velocity and the flow velocity.

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