Effect of Pressure on the Lean Limit Flames of H₂-CH₄-Air Mixture in Tubes

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Abstract

The lean limit flames of H₂-CH₄-air mixtures stabilized inside tubes in a downward flow are experimentally and numerically investigated at elevated pressures ranging from 2 bar to 5 bar. For the shapes of lean limit flames, a change from ball-like flame to cap-like flame is experimentally observed with the increase of pressure. This experimentally observed phenomenon is qualitatively predicted by numerical simulations. The structure of ball-like and cap-like lean limit flames at all tested pressures is analysed in detail based on the numerical predictions. The results show that the lean limit flames are located inside a recirculation zone at all tested pressures. For the leading edges of the lean limit flames at all tested pressures, the fuel transport is controlled by both convection and diffusion. For the trailing edge of the ball-like lean limit flame at 2 bar, the fuel transport is dominated by diffusion. However, with increasing pressure, the transport contribution caused by convection in the trailing edges of the lean limit flames increases. Finally, the influence of transport and chemistry on the predicted ultra lean flames and lean flammability limit is analysed at elevated pressures.

Keywords: Ultra Lean, Ball-like Flame, Cap-like Flame, Low Lewis Number

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1. Introduction

Fundamental understanding of ultra lean flame combustion characteristics is essential for fire safety control as well as for the development of high-efficiency low-emission engines based on ultra lean combustion concepts. The so-called flame ball [1] is a type of flame structure that can form in ultra lean low Lewis number mixtures [2]. In fact, at micro-gravity conditions a transition from cell-like flame to flame ball occurs as the equivalence ratio approaches the lean limit [2, 3, 4], and the lean limit is extended to much leaner mixtures. Studies on flame balls assist to improve our understanding of transport and chemistry properties of the ultra lean low Lewis number flames, and extended flammability limits.

Zeldovich [1] first theoretically predicted that a purely diffusion controlled flame with a steady radius could exist. However, he pointed out that adiabatic flame ball solutions are unstable, and further hypothesized that a flame ball could be stabilized by heat loss. Ronney [2] accidentally observed apparent flame balls at micro-gravity conditions for a ultra lean H_2 -air mixture in a drop tower experiment. Subsequent aircraft-based micro-gravity experiments [3] showed that flame balls can exist in different near-limit low Lewis mixtures of H_2 -air, H_2 - O_2 , H_2 - O_2 - SF_6 , and CH_4 - O_2 - SF_6 . Also, space shuttle experiments [4] showed that multiple flame balls experience low and repulsive drift speeds. Recently, Takase et al. [5] reported that a ball-like low Lewis number flame exists near the lean flammability limit in a convective flow at micro-gravity conditions.

The effects of radiative heat loss on flame balls were studied by Buckmaster et al. [6, 7], concluding that flame balls become stable with the inclusion of radiative heat loss. Comprehensive one-dimensional numerical simulation of non-adiabatic flame balls [8, 9] has been conducted with detailed chemistry, transport and radiation models, demonstrating that numerical results are qualitatively consistent with the experimental results. Kagan and Sivashinsky [10] and Brailovsky and Sivashinsky [11] numerically and theoretically predicted that a flame ball is able to exist in the convective flow (so-called travelling flame ball), indicating that propagating flame balls may be obtained in experiments.

While all the above studies were conducted at micro-gravity conditions or zero-gravity conditions, Shoshin and de Goeij [12] observed a transition from cell-like propagating flames to ball-like

propagating flames of $\text{H}_2\text{-CH}_4\text{-air}$ mixtures near the lean limit in experiments on flame propagation in a tube at normal gravity. These propagating ball-like flames were further experimentally and numerically investigated by Shoshin et al. [13]. Subsequently, stationary ball-like flames in $\text{H}_2\text{-CH}_4\text{-air}$ mixtures near the lean limit with well defined boundary conditions were stabilized in a downward flow [14] at normal pressure. A detailed comparison between experimental ball-like and numerical ball-like flames at normal pressure has been conducted by Hernández-Pérez et al. [15], analyzing the effects of transport, chemistry and heat loss on the formation and stabilization of the ball-like flames.

To our knowledge, not much research has been published about the lean limit flames with low Lewis number at elevated pressures, despite the fact that combustion in practical devices (e.g., gas turbines and engines) usually occurs at elevated pressures. Furthermore, H_2 is considered as fuel for power generation utilizing heavy duty gas turbines [16]. Therefore, it is of great interest to understand the influences of pressure on the lean flammability limit and low Lewis number lean limit flame behaviour.

The aim of this work is to experimentally and numerically investigate lean limit flames with low Lewis number at elevated pressures and normal gravity. In order to keep the consistency with the previous study at normal pressure [15], a 40% H_2 and 60% CH_4 fuel composition (specified on a molar basis) is selected. Lean limit flames at elevated pressures are experimentally and numerically examined as a function of the inlet equivalence ratio, ϕ . The lean limit flames are analysed in detail using the numerical solutions. In the following sections, the experimental setup and numerical approach are presented, and the experimental and numerical results are discussed. Finally, the concluding remarks are drawn.

2. Experimental setup

An experimental setup similar to the one used in [15] is employed in this study. In order to extend this setup to elevated pressures, it is redesigned to fit in the high pressure cell which was used in [17]. A schematic of the experimental setup is shown in Fig. 1. A laminar flame is stabilized inside a cylindrical silica tube. Three gas lines are used to supply the hydrogen-methane, air and cooling air, controlled by mass flow controllers with an uncertainty of less than 1%. A

Figure 1: Schematic of high pressure flame ball setup.

buffer is mounted to damp the small flow fluctuation before the mixture is supplied to the burner. Also, a perforated plate with a thickness of 2 mm and holes of 0.5 mm diameter uniformly spaced with 0.3 mm pitch is mounted at the top of the tube to generate a uniform inlet flow profile. The unburned mixture at a sufficiently large equivalence ratio is ignited by a spark plug that is installed under the burner. Then the equivalence ratio is step-wisely reduced to the lean flammability limit. The temperature of tube wall is kept nearly constant by supplying cooling air through a co-axial slit situated above the tube. Three K-type thermal-couples are mounted into small holes with a depth of around 0.6 mm in the tube wall in order to accurately measure the temperature of the inner tube wall. The measured tube wall temperatures in all the experiments are in the range of 300-320 K for the three locations. A pressure sensor is installed to measure the pressure inside the cell, which is further connected to a pressure regulator.

CH^* chemiluminescence of the flames is recorded by an AVT-PIKE F-032b CCD-camera equipped with an interference filter (430 nm and bandwidth 10 nm). The chemiluminescence

images show a line-of-sight integrated emission intensity. The in-plane radial emission intensity distribution is recovered by Abel inversion for the cylindrically symmetric flames. In order to investigate the tube diameter effect on the formation of lean limit flames, two different fused silica tubes are employed in the experiments, having the following geometrical specifications: (a) 30 mm height, 13.5 mm inner diameter, 1 mm wall thickness and (b) 30 mm height, 7 mm inner diameter, 1 mm wall thickness. The experimental conditions are summarized in Table 1, which also includes the experimental condition for ball-like flame at normal pressure [15] for reference. The inlet velocities in the experiments at all tested pressures are chosen near the blow off limit for the lean limit flames. Equivalence ratio is decreased until the lean limit is reached.

Table 1: Summary of conditions for present experiments. The experimental condition for ball-like flame at normal pressure [15] is also included for reference. P is the initial pressure, D_i is the tube inner diameter and V_{in} is the inlet velocity.

Condition	P (bar)	D_i (mm)	V_{in} (cm/s)
I	1	13.5	7.2
II	2	13.5	8.0
III	2	7	5.0
IV	3	7	5.8
V	4	7	6.0
VI	5	7	6.2

3. Numerical approach

Detailed numerical simulations of the flames are performed with the body-fitted, multi-block, adaptive mesh refinement, finite-volume framework that has been originally developed by Groth and co-researchers [15, 18, 19]. In the present research work, the conservation equations of mass, momentum, energy and species are solved in a cylindrical coordinate system. The full equations can be found in our previous publication [15]. The flow is treated as a mixture of perfect gases under the assumption of an ideal gas equation of state. Thermodynamic properties, transport properties and species net production/destruction rate are all computed by the open-source library CANTERA [20].

In the simulations, the computational domain is a cylindrical channel with a radius of 3.5 mm and a height of 30 mm, as shown in Fig. 2. Because the flames in this study are axisymmetric, only half two-dimensional (2D) equivalent flames are simulated. 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 which accounts for the diffusion of fuel/air mixture across the upper boundary. The fuel/air mixture is supplied uniformly from the upper boundary. The right boundary is treated as a no-slip and iso-thermal boundary with a temperature of 300 K according to the experimental measured tube wall temperature (less than 320 K) in the experiments. The lower boundary is modelled as a free outflow.

Two different diffusion models are utilized in order to assess the transport effect on the prediction of lean limit flames. The diffusion velocities are computed with the Mixture-averaged (MAV) and Multi-Component with Soret diffusion (MCS) transport models in the simulations, which are given by

$$\text{MAV} : U_i = -\frac{D_{im}}{X_i} \nabla X_i \quad (1)$$

$$\text{MCS} : U_i = -\frac{1}{X_i \bar{W}} \sum_{j \neq i}^M W_j D_{i,j} \nabla X_j - \frac{D_i^T}{\rho Y_i} \frac{1}{T} \nabla T \quad (2)$$

where D_{im} is the mixture-averaged diffusion coefficients of species i , X_i is the molar fraction of species i , W_j is the molar mass of species j , $\bar{W}$ is the mean molar mass of the mixtures, $D_{i,j}$ is the binary multicomponent diffusion coefficients for species i and j , ρ is the density, and D_i^T is the thermal diffusion coefficient of species i . The GRI 3.0 chemical mechanism [21] without inclusion of NO_x sub-mechanism consisting of 36 species with 219 reactions is employed in this study. A skeletal mechanism [22] for CH_4 which consists of 16 species and 25 elementary reactions is also employed in four of the cases for comparison with the GRI 3.0 chemical mechanism. Gravity and radiative heat loss are taken into consideration. Thermal radiation is modelled via the optically-thin Planck model. The Planck mean absorption coefficient of the mixture is evaluated from the major radiating species CO_2 , H_2O and CO . The individual coefficients are calculated using a statistical

Figure 2: Schematic of the computational domain and boundary conditions.

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 $44\ \mu\text{m}$ for various pressures. These levels of resolution are sufficient to get grid-independent solutions for all tested pressures due to the rapid increase of flame thickness for the near lean limit flames. The thermal thicknesses of the ball-like flames at different pressures in 7 mm diameter tube are about 2 mm. In this case, there are about 45 grid points within the flame thickness. More details of the numerical method can be found in [15]. The simulated cases are listed in Table 2. It shows that, for a fixed equivalence ratio, pressure has no influence on the adiabatic flame temperature. Mixture-averaged transport model and GRI 3.0 chemical mechanism are employed in the simulation for qualitative prediction of experimentally measured flames near lean flammability limit, due to the extremely expensive computation time using Multi-component transport model with Soret diffusion and GRI 3.0 chemical mechanism. The influence of transport model and chemistry on the predicted lean limit flames will be examined in section 4.5.

Table 2: Summary of the simulated cases with a tube inner diameter of 7 mm using Mixture-Averaged transport Model and GRI 3.0 Mechanism [21]. T_{ad} is the adiabatic flame temperature.

Case	P (bar)	V_{in} (cm/s)	ϕ	T_{ad} (K)
A	2	5.0	0.45	1405
B	2	5.0	0.40	1302
C	2	5.0	0.38	1260
D	2	5.0	0.37	1239
E	3	5.8	0.45	1405
F	3	5.8	0.40	1302
G	3	5.8	0.37	1239
H	3	5.8	0.35	1196
I	4	6.0	0.40	1302
J	4	6.0	0.37	1239
K	4	6.0	0.35	1196
L	4	6.0	0.345	1185
M	5	6.2	0.40	1302
N	5	6.2	0.37	1239
O	5	6.2	0.35	1196
P	5	6.2	0.348	1191

4. Results and discussion

4.1. Effect of tube diameter on the formation of lean limit flames

Since a ball-like lean limit flame was observed in a tube with an inner diameter of 13.5 mm at normal pressure in ref. [15], the experiments in the present study are initially conducted with this tube at 2 bar. Figure 3 shows the CH^* chemiluminescence distributions of the flames stabilized inside the 13.5 mm diameter tube at 2 bar with different equivalence ratios. The CH^* chemiluminescence distributions of the flames inside this tube at 1 bar with different equivalence ratios are also plotted in Fig. 3 for reference. Cell-like flames are observed for equivalence ratios far away from the lean limit at 2 bar. However, as shown in Fig. 4(a), the flame takes a disc-like shape with a relatively bright edge at an equivalence ratio of 0.33. With further slight decrease of equivalence ratio to $\phi = 0.325$, the middle part of the flame becomes weaker and then the flame breaks into two flame-lets which move chaotically. Later these ball-like flamelets are extinguished by the convective flow. This phenomenon is somehow similar to the near lean limit flame behaviour reported in [12]. This could be due to the combined effects of the hydrodynamic instability and

Figure 3: The Abel inverted CH^* chemiluminescence images of the flames studies in the experiments with the tube diameter of 13.5 mm at 2 bar (Upper) and 1 bar (Lower).

thermal-diffusive instability. The hydrodynamic instability increases with the increase of pressure and the thermal-diffusive instability increases with the decrease of equivalence ratio, which leads to amplification of small perturbations. The size of the drifting ball-like flamelet (Fig. 4(d)) at 2 bar is significantly smaller than that at 1 bar. Therefore, in order to obtain a stable ball-like flame at 2 bar, a tube with an inner diameter of 7 mm is selected for the experiments at elevated pressures after trying different tubes with different tube diameters. The remainder of this paper only focuses on flames inside a tube with an inner diameter of 7 mm.

For a more rigorous analysis on the selection of tube diameter at 2 bar, the cut-off wavelengths for hydrodynamic instability and thermal-diffusive instability are estimated through classical dispersion relations [24, 25]. The estimated cut-off wavelengths for hydrodynamic instability and thermal-diffusive instability are approximately $20\delta_f$ and $8\delta_f$, respectively. δ_f is the flame thick-

Figure 4: Transient images of the flames studied in a tube with a diameter of 13.5 mm at 2 bar after a decrease of ϕ from 0.33 to 0.325.

ness, which is usually evaluated using the following equation:

$$\delta_f = \frac{\lambda}{\rho V C_p} \quad (3)$$

where λ is the heat conductivity, V is the flame propagation speed, and C_p is the specific heat at constant pressure. However, the flame propagation speed for the cases in the present study cannot be estimated, because the lean limit flames are located inside a recirculation zone. Therefore, the thermal thicknesses for the ball-like lean flames along the flame centerline at 1 bar and 2 bar are calculated based on the numerical gradients of temperature, yielding: 3.51 and 1.95 mm, respectively. Although the estimated cut-off wavelengths for hydrodynamic instability and thermal-diffusive instability are larger than the corresponding tube diameter, the ratio of thermal flame thicknesses at 1 bar and 2 bar are very close to the corresponding ratio of tube diameters. This implies that the ball-like lean limit flames form when the ratio of flame thickness and tube diameter is closer to a certain value.

Figure 5: The Abel-inverted CH^* chemiluminescence images of the flames studied in the experiments with a tube diameter of 7 mm for the pressures ranging from 2 bar to 5 bar.

4.2. Flame shapes at different pressures

Figure 5 shows the Abel inverted CH^* chemiluminescence distributions of the flames in the 7 mm diameter tube with different equivalence ratios at all four tested pressures. It is seen that a more spherical ball-like flame is obtained inside the tube at 2 bar as the lean limit is approached, as compared to 1 bar in ref. [15]. However, with the increase of pressure, for the lean limit flames, a change from ball-like flames to cap-like flames is observed. This phenomenon is similar to the findings in ref. [12] being that a change from ball-like lean limit flame to cap-like lean limit flame occurs with the increase of tube diameter. This indicates that the change of combustion regime in tubes could depend on a key factor that can be impacted by both tube diameter and pressure.

Numerical simulations are performed on these lean limit flames at all tested pressures with the Mixture-averaged transport model and the GRI-Mech 3.0 chemical mechanism. Figure 6 displays the distributions of chemical heat release and streamlines at all tested pressures for different

equivalence ratios. It can be seen that qualitatively the numerical predictions are able to predict the transition from cell-like flame to ball-like flame at 2 bar and to cap-like flames at the higher pressures with the decrease of equivalence ratio. Figure 6 further shows that recirculation zones occur in the hot core region for the flames near the lean flammability limit due to the buoyancy force acting on the hot gases in the flame region. Furthermore, the size and the intensity of recirculation rapidly increase with the increase of pressure, which leads to a less curved trailing edge of the lean limit flames at higher pressures. This is because the impact of buoyancy on the lean limit flames increases with increasing pressure. At the same time, the increase in buoyancy effect also leads to a higher propagation speed for lean limit flames at higher pressures, because the propagating speed for the lean limit flames in tubes is determined by buoyancy [12]. Therefore, the increase in inlet velocity near the blow-off limit in the experiments with the increase of pressure can be explained by the increase in buoyancy effect at higher pressures. Furthermore, with higher inlet velocity, the heat loss caused by heat conduction to the tube wall decreases, as shown in Table 3. Consequently, the lean limit flame spreads out in the tube with a rise of pressure. To sum up, nearly spherical ball-like lean limit flames tend to form at conditions with less impact of buoyancy. This can be achieved by reducing the tube diameter at elevated pressures.

To further compare the flame shapes from the experiments with those from the 2D numerical predictions, the half-height and half-width of the flames are plotted in Fig. 7 as a function of the equivalence ratio. The peaks of CH^* chemiluminescence for the left and right parts and the upper and bottom parts of all the flames in the experimental images and the peaks of heat release for the numerical simulations are used to determine the flame characteristic lengths. At 2 bar, the height and the width of the flames for both experiments and numerical simulations converge to nearly the same value with decreasing equivalence ratio. The leanest flame in the experiments is at $\phi = 0.31$. However, the numerical solution of the leanest flame is obtained at $\phi = 0.37$. Notably, the discrepancies of the lean flammability limits obtained by the experiments and the numerical simulations and the characteristic lengths extracted from the experiments and the numerical simulations diminish with the increase of pressure. The reason for the discrepancy between experimental lean limit and numerical lean limit may be due to the combined effects of heat loss, transport and chemistry. The influences of transport and chemistry on the lean flammability limit will be discussed in

section 4.5.

It is also of interest to compare the measured ball-like flame size at 2 bar with the 1D micro-gravity flame ball radius. It should be noted that, for 1D micro-gravity flame balls, the heat loss is caused by radiation. However, for ball-like lean limit flame in the tube, the heat loss is dominated by convection and heat conduction to the tube wall rather than radiative heat loss, as shown in Table 3. The different main heat loss mechanism could lead to different flame size and lean limit between the 1D micro-gravity flame balls and 2D ball-like flames. Therefore, for comparison, the 1D micro-gravity flame balls with different radiative heat loss are simulated using the in-house code CHEM1D [26] using the same chemistry and transport models. The radiative heat loss is varied by an arbitrarily increase of the calculated Planck mean absorption coefficient by factors 1.8 and 2.6. Multi-component transport model with the inclusion of Soret diffusion, GRI-mech 3.0 chemical mechanism and optically thin radiative model are employed in the 1D simulation. The details of the 1D simulation are described in [9, 15]. The measured ball-like flame size and 1D micro-gravity flame ball radius with different radiative heat losses are plotted in Fig. 8. It is seen that, with the increase of radiative heat loss, the radii of 1D micro-gravity flame balls decrease and the lean limit equivalence ratio of 1D micro-gravity flame ball increases. The 1D micro-gravity flame ball radii and lean flammability limit with the 2.6 times enhanced Planck mean absorption coefficient are in agreement with the half-width and lean flammability limit of the measured ball-like flame. This result implies that combustion mechanisms of the ball-like flames in present work and the micro-gravity flame balls could be similar, while discrepancy for sizes and lean limits between the two types of flames can be explained by the difference in the value of heat loss. Additionally, Table 3 also shows that conductive heat loss becomes less important while the influence of radiative heat loss increases at higher pressures. Furthermore, the leakage of fuel increases at higher pressures.

Table 3: Energy in fuel stream (Q_{fu}), integrated heat release (Q_{chem}), conductive heat loss to the tube wall from upwind maximum heat release to downwind maximum heat release (Q_{cond}) and radiative heat loss (Q_{rad}) from the 2D predicted lean limit flames at the pressures between 2 bar and 5 bar.

P (bar)	ϕ	Q_{fu} (W)	Q_{chem} (W)	Q_{cond} (W)	Q_{rad} (W)
2	0.370	4.21	1.13	0.155	0.023
3	0.350	7.74	1.62	0.095	0.043
4	0.345	10.55	1.96	0.062	0.064
5	0.348	13.70	2.17	0.051	0.080

Figure 6: Distribution of chemical heat release (W/m^3) and flow streamlines of the flames studied in the simulations with mixture-averaged transport and GRI-mech 3.0, corresponding to the various pressures in the experiments. (a-d): 2 bar; (e-h): 3 bar; (i-l): 4 bar; (m-p): 5 bar.

Figure 7: Half-heights and half-widths from the 2D numerical predictions and experiments as a function of equivalence ratio at all tested pressures.

Figure 8: The radius of 1D micro-gravity flame ball with different heat losses and the characteristic lengths of the flames obtained from the experiments as a function of equivalence ratio at 2 bar.

4.3. Flame structure at different pressures

In Fig. 9, the distributions of the normalized heat release, the normalized temperature and the scaled mass fraction of species along the centerline for different equivalence ratios at all tested pressures are presented. The mole fraction of species is scaled by the maximum mole fraction of H_2O . It can be seen in Fig. 9 that the mole fractions of H_2 and CH_4 first decrease and then increase from the inlet to outlet for all the tested cases. This is because the unburned mixture leaks from the upstream side to the downstream side following the streamlines near the tube wall. It further shows that the leakage of the unburned mixture increases with the decrease of equivalence ratio due to the reduction in flame size. This leads to an enhancement of the heat release of the trailing edge with the decrease of equivalence ratio at all tested pressures. Meanwhile, the peak values of the heat release at the centerline come closer to each other with the decrease of equivalence ratio. In addition, for the lean limit flames, the heat release of the trailing edge relative to the heat release of the leading edge at the centerline decreases with the increase of pressure. This is because less fuel diffuses to the centerline due to the increase of the flame width with increasing pressure.

As the micro-gravity flame balls are exclusively controlled by diffusion, it is interesting to show the convective and diffusive fluxes of CH_4 and H_2 for the 2D lean limit flames at different pressures to access inheritance of diffusive transport in normal gravity ball-like flame combustion mechanism. As a first step, for the comparison with the 2D lean limit flames, the convective and diffusive fluxes of CH_4 and H_2 for two 1D flat flames ($\phi = 0.55$) at 2 bar and 5 bar and two 1D micro-gravity flame balls near the lean limit ($\phi = 0.31$) at 2 bar and 5 bar are computed. The results are plotted in Fig. 10 as a function of normalized temperature, $c = (T - T_u)/(T_b - T_u)$, where T_u and T_b are the unburned mixture temperature and maximum flame temperature. For flat flames, a convection dominated preheat zone and a diffusion dominated reaction zone are observed. The convective flux of CH_4 decreases linearly with the increase of temperature in the preheat zone while the diffusive flux of CH_4 increases linearly. However, the convective flux of H_2 decreases significantly in the initial stage of the increase in temperature while the diffusive flux of H_2 increases greatly. This is due to the high diffusivity of H_2 . Additionally, the diffusive fluxes of CH_4 and H_2 are both large in the reaction zone for the 1D flat flame. Furthermore, both convective and diffusive fluxes increase with increasing pressure. However, the micro-gravity

Figure 9: Normalized Heat Release, normalized temperature and scaled mole fraction of species along the centerline of predicted flames for different equivalence ratios at all tested pressures.

flame balls are completely controlled by diffusion since convection is absent, as shown in Fig. 10(c, d). Furthermore, the diffusive flux of H₂ dominates that of CH₄ and the diffusive fluxes increase

Figure 10: The convective and diffusive fluxes of CH_4 and H_2 for 1D flat flames ($\phi = 0.55$) at 2 bar and 5 bar and 1D micro-gravity flame balls ($\phi = 0.31$) at 2 bar and 5 bar.

significantly with the increase of pressure.

Now turning to normal gravity ball-like flames, Figure 11 displays the distributions of the convective and diffusive fluxes of CH_4 and H_2 for the leading edges at the centerline of the tube as a function of normalized temperature for different equivalence ratios at 2 bar and for the limit flames at different pressures. It is seen that the convective fluxes of both CH_4 and H_2 decrease significantly in the initial stage of the increase in temperature for all the cases and more so at lower equivalence ratios. This reflects that the fuel transport caused by the convection for the leading edges plays a less important role approaching the lean limit, as compared with that in the 1D flat lean limit flame. Furthermore, unlike the diffusive fluxes of CH_4 and H_2 in the 1D flat flame, the diffusive flux of H_2 for the leading edges dominates that of CH_4 , which is similar to the distributions of the 1D micro-gravity flame ball (Fig. 10). This further indicates that convective transport for the leading edges is less important than that for the 1D flat lean limit flame. The

Figure 11: The convective and diffusive fluxes of CH_4 and H_2 for the leading edges at the centerline of the tube as a function of normalized temperature for different equivalence ratios at 2 bar (Upper) and for lean limit flames at different pressures (Lower).

Transport in the major part of the preheat zone ($c > 0.25$) for all the lean limit flames at different pressures is dominated by diffusion. Besides, both convective and diffusive fluxes increase with increasing pressure for all simulated cases.

Figure 12 displays the distributions of the convective and diffusive fluxes of CH_4 and H_2 for the trailing edges at the centerline of the tube as a function of normalized temperature for different equivalence ratios at 2 bar and for the lean limit flames at different pressures. It is observed that the diffusive fluxes of CH_4 and H_2 increase with the decrease of equivalence ratio as for the leading edge. The increase is because more unburned mixture leaks from the upstream side to the downstream side with the decrease of equivalence ratio. Due to higher diffusivity of H_2 , the diffusive flux of H_2 dominates that of CH_4 similar as found for the leading edge and for 1D flame balls. At 2 bar, the convective fluxes of CH_4 and H_2 are negative, reflecting that CH_4 and H_2 are transported out of the trailing edge by convection. Therefore, the trailing edges at 2 bar are

Figure 12: The convective and diffusive fluxes of CH_4 and H_2 for the trailing edges at the centerline of the tube as a function of normalized temperature for different equivalence ratios at 2 bar (Upper) and for lean limit flames at different pressures (Lower).

dominated by diffusion. With the increase of pressure, the convective fluxes of CH_4 and H_2 change from negative values to positive values for the lean limit flames, indicating that the trailing edges of the lean limit flames are more affected by the convective flow at higher pressures. This is due to the enhancement of the recirculation zone intensity at higher pressures. It can also be seen in Fig. 11 and 12 that the diffusion flux curves for ball-like lean limit flames at 2 bar and 3 bar closely resemble the one found for 1D micro-gravity flame balls (Fig. 10). However, for above-limit flames at 2 bar and cap-like lean limit flames at 4 bar and 5 bar, the diffusive flux profiles deviate more significant from the one found for 1D flame balls.

Figure 13: Maximum $(\phi_{local} - \phi_{inlet})/\phi_{inlet}$ with different equivalence ratio at all tested pressures.

4.4. Effect of preferential diffusion on lean limit flames

For flames with low Lewis number, the local equivalence ratio can be significantly changed by the coupling effects of preferential diffusion and flame stretch/curvature. In this part, the local equivalence ratio is calculated based on the definition in ref. [27], which is given by

$$\phi_{local} = \frac{0.5(X_{H_2} + X_{H_2O}) + X_{CO_2} + X_{CO} + 2X_{CH_4}}{0.5(X_{CO} + X_{H_2O}) + X_{O_2} + X_{CO_2}} \quad (4)$$

where X_i is the mole fraction of the major species which are considered in the calculation. The maximum of the quantity, $(\phi_{local} - \phi_{inlet})/\phi_{inlet}$, are presented in Fig. 13 for different inlet equivalence ratios for all tested pressures. It shows that the maximum increment of the equivalence ratio in the flame increases with the decrease of equivalence ratio. This can be explained by the increase in flame curvature as equivalence ratio decreases. Furthermore, at 2 bar, the maximum change increases slightly more rapidly as the equivalence ratio approaches the lean limit, as compared with at higher pressures which can be attributed to the larger curvature of near-limit flames at 2 bar compared to the flames at higher pressures.

The quantity, $(\phi_{local} - \phi_{inlet})/\phi_{inlet}$, along the centerline of the leanest flames are plotted in Fig. 14 as a function of normalized temperature, c , for all tested pressures. As shown in Fig. 14, for the leading edges of the flames, the local equivalence ratio first decreases slightly relative to inlet equivalence ratio and then increases in the flame front. This is because H_2 diffuses faster to the

Figure 14: Distributions of the $(\phi_{local} - \phi_{inlet})/\phi_{inlet}$ and $(C/H_{local} - C/H_{inlet})/(C/H_{inlet})$ at the centerline of the tube as a function of normalized temperature (c) for the lean limit flames at all tested pressures.

reaction zone than O_2 . For the trailing edges of the flames, the local equivalence ratio increases almost monotonously with the increase of temperature. No remarkable difference is observed for the $(\phi_{local} - \phi_{inlet})/\phi_{inlet}$ at different pressures except that the $(\phi_{local} - \phi_{inlet})/\phi_{inlet}$ at 2 bar is slightly lower than that at higher pressures.

4.5. Effect of transport model and reaction mechanism on lean limit flames

To examine the influence of transport and chemistry on the predicted lean limit flames, four additional cases (R, Q, S and T) are simulated with an equivalence ratio of 0.37 at 2 bar and 5 bar, as shown in Table 4 with inclusion of two reference cases (D, N). Distributions of chemical heat release rate simulated with different transport models and chemical mechanisms are displayed in Fig. 15. It can be seen that, at 2 bar, the predicted flame of case R has a larger size than that of case Q. Furthermore, the predicted flame of case D has a slightly smaller size than that of case Q. This indicates that, at 2 bar, the Multi-component transport model with Soret diffusion is able to predict much leaner flame than the Mixture-averaged transport model. The difference between experimental and numerical lean limits is expected to be reduced when the more accurate Multi-component transport model with Soret diffusion is employed in the simulation.

At 5 bar, the leanest flame predicted with the skeletal chemical mechanism is obtained at $\phi = 0.37$ using the Mixture-averaged transport model, while for the GRI-3.0 mechanism it is at $\phi = 0.346$ using the same transport model. The latter value is in agreement with the lean limit in the experiment. However, the predicted flame of case T still has a larger size than that of case S. This implies that the leanest flame predicted with the Multi-component transport model with Soret diffusion and GRI-3.0 mechanism can reach much leaner equivalence ratio than in experiment. The lean limit flames at elevated pressures are very sensitive to the transport and chemistry model. Caution has to be taken for the choice of transport model and chemical mechanism at elevated pressures.

Table 4: Summary of four additional simulated cases for flames in a 7 mm diameter tube with a skeletal mechanism for CH₄ [22] using different transport models. MAV is the Mixture-Averaged Model and MCS is the Multi-Component Model with Soret diffusion. Two reference cases (D, N) are also included.

Case	P (bar)	V_{in} (cm/s)	ϕ	Transport Model	Chemical Model
D	2	5.0	0.37	MAV	GRI 3.0
N	5	6.2	0.37	MAV	GRI 3.0
Q	2	5.0	0.37	MAV	Skeletal
R	2	5.0	0.37	MCS	Skeletal
S	5	6.2	0.37	MAV	Skeletal
T	5	6.2	0.37	MCS	Skeletal

Figure 15: Distributions of chemical heat release of the flames simulated with different transport models and chemical mechanisms, corresponding to $\phi = 0.37$ at $P = 2$ bar and 5 bar. (a,d): Mixture-averaged transport model and GRI-mech 3.0; (b,e): Mixture-averaged transport model and Skeletal Mechanism; (c,f): Multi-component transport model with Soret diffusion and Skeletal Mechanism. (a-c): 2bar; (d-f): 5 bar.

To further quantify the influence of transport and chemistry, the consumption rate of H_2 and CH_4 is integrated over the entire domain for the cases (D, Q, R, N, S and T). The integrals of consumption rate for the cases predicted with Multi-component transport model with Soret diffusion and skeletal chemical mechanism (R, T) and with Mixture-averaged transport model and GRI-3.0 mechanism (D, N) are scaled by the corresponding cases predicted with Mixture-averaged transport model and skeletal chemical mechanism (R, S). The results are presented in Table 5. The scaled consumption rates of H_2 and CH_4 for 1D flat flames with an equivalence ratio of 0.55 at 2

bar and 5 bar are included as well. It shows that, at 2 bar, for both 2D flames and 1D flat flames the GRI-mech 3.0 predicts slightly lower consumption rates for both H_2 and CH_4 than the skeletal mechanism. However, Soret diffusion increases the consumption rates of H_2 and CH_4 by factors 1.38 and 1.43 for 2D flames respectively, while the Soret diffusion has almost no influence on the consumption rate of the 1D flat flames. This is because, for 2D flames, the Soret diffusion effect is enhanced by the flame curvature. With the increase of pressure from 2 bar to 5 bar, the GRI-mech 3.0 predicts larger consumption rates for both H_2 and CH_4 than the skeletal mechanism for the 2D flames, while for 1D flat flames the GRI-mech 3.0 predicts only slightly larger consumption rate. Furthermore, the influence of Soret diffusion on the consumption rate of the 2D flames increases with the increase of pressure. To sum up, the numerical prediction of lean limit flames becomes more sensitive to the transport and chemistry models with the increase of pressure. At low pressure, the discrepancies between experimental and numerical results can be significantly reduced by employing multi-component transport model, while, at high pressure, these discrepancies increase when the multi-component transport model is utilized. A good agreement between experimental results and numerical results based on the mixture-averaged transport model and GRI 3.0 mechanism at 5 bar just happens for this specific case by coincidence. This actually implies that the uncertainties of the chemical mechanism at ultra lean conditions are large at high pressures.

Table 5: Normalized consumption rates of H_2 and CH_4 at 2 bar and 5 bar.

Cases	P (bar)	Factor	2D		1D	
			H_2	CH_4	H_2	CH_4
R/Q	2	Soret diffusion	1.38	1.43	1.01	1.01
D/Q	2	Chemistry	0.97	0.89	0.89	0.89
T/S	5	Soret diffusion	1.5	1.58	1.01	1.01
N/S	5	Chemistry	1.46	1.72	1.14	1.14

5. Conclusions

The influences of pressure on the lean limit H_2 - CH_4 -air flames in tubes are experimentally and numerically investigated for a fuel with 40% H_2 and 60% CH_4 (on a molar basis) premixed with air. A ball-like lean limit flame is experimentally stabilized by downward flow at 2 bar. With a further increase of pressure, a change from ball-like lean limit flame to cap-like lean limit flame is observed. Numerical simulations are performed with a mixture-averaged transport model and GRI-Mech 3.0 to investigate the ball-like and cap-like lean limit flames at all tested pressures. A good qualitative agreement between predicted flames and experimental flames is observed.

Detailed analysis of the flame structure is conducted for the lean limit flames at all tested pressures based on the predicted solutions. It is found that the lean limit flames are located inside a recirculation zone. The size of the recirculation zone for the lean limit flames increases with increasing pressure. For the leading edges of the lean limit flames, a flame structure with negative displacement speed is observed. As compared with the convective and diffusive fluxes in a 1D flat flame, fuel transport caused by the convective flux in the leading edges of the lean limit flames is less important. For the trailing edge of the ball-like flame at 2 bar, transport is mainly controlled by diffusion. With an increase of pressure, fuel transport caused by the convective flux in the trailing edge increases. Ball-like lean limit flames tend to form inside the tube at conditions with less influence of buoyancy, which can be achieved by reducing tube diameter at elevated pressures. The transition from ball-like to cap-like lean limit flames with increasing pressure is caused by buoyancy.

Additionally, the influence of transport and chemistry on the predicted ultra lean flames and lean flammability limit at 2 bar and 5 bar has been examined. It shows that the discrepancy between experiments and numerical simulation is mainly caused by the simple transport model at 2 bar. However, the prediction of lean limit flames is sensitive to both chemistry and transport at higher pressures. This makes the current experimental platform important not only for studying Low Lewis lean limit flames but also a suitable setup for the development and validation of reaction mechanisms and transport models at ultra lean conditions.

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