Title: High-speed imaging of OH and acetone PLIF reveals the structure of methane inverse diffusion flames

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Keywords: inverse methane diffusion flame; high speed imaging; flame structure.

Abstract: Most previous work on inverse diffusion flames (IDFs) has focused on laminar IDF emissions and the soot formation characteristics. Here, we investigate the characteristics and structure of methane IDFs using high speed planar laser-induced fluorescence (PLIF) images of OH, particle image velocimetry (PIV), and acetone PLIF imaging for non-reacting cases. First, the flame appearance was investigated with fixed methane loading (mass flux) but with varying airflow rates, yielding a central air jet Reynolds number (Re) of 1,000 to 6,000 (when blow-off occurs). Next, it was investigated a fixed central air jet Re of 4500, but with varied methane mass flux such that the global equivalence ratio spanned 0.5 to 4. It was observed that at Re smaller than 2000, the inner air jet promotes the establishment of an inverse diffusion flame surrounded by a normal diffusion flame. However, when the Re was increased to 2500, two distinct zones became apparent in the flame, a lower entrainment zone and an upper mixing and combustion zone. 10 KHz OH-PLIF images, and 2D PIV allow the identification of the fate and spatial flame structure. Many flame features were identified and further analyzed using simple but effective image processing methods, where three types of structure in all the flames investigated here: flame holes or breaks; closures; and growing kernels. Insights about the rate of evolution of these features, the dynamics of local extinction, and the sequence of events that lead to re-ignition are reported here. In the lower entrainment zone, the occurrence of the flame break events is counterbalanced by closure events, and the edge propagation appears to control the rate at which the flame holes and closures propagate. The rate of propagation of holes was found to be statistically faster than the rate of closure. As the flames approach blow-off, flame kernels become the main mechanism for flame re-ignition further downstream. The simultaneous OH-PLIF/Stereo PIV measurements indicate that the individual breaks events could be correlated to local vortical flow structure and strain rates fields. The detailed measurements provide a more complete understanding of IDF flame characteristics and structure than was previously possible.
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I am enclosing here with a manuscript entitled “High-speed imaging of OH and acetone PLIF reveals the structure of methane inverse diffusion flames” for evaluation. With the submission of this manuscript, I would like to undertake that the above mentioned manuscript has not been published elsewhere, accepted for publication elsewhere or under editorial review for publication elsewhere.

Sincerely yours,

Ayman elbaz on behalf of the authors

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Dear Prof. John Patrick:

Thank you for accepting revising our paper and we appreciate the efforts of the reviewers. They have carefully critiqued our manuscript and provided important comments to improve the work. We have considered all their comments and responded by modifying the manuscript. We believe the revised manuscripts is a considerable improvement and suitable for publication in FUEL.
In the attached pages, we indicate the changes to the paper in response to the reviewers’ comments. The reviewers’ comments are given in red italic and our response is directly following. The changes in the manuscript are indicated in underline red text.

I appreciate your efforts with regard to the manuscript.

Best Regards,
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Response to Reviewer 2:
Reviewer #2: It is grateful to note that the authors have responded to all the reviewers’ comments and revised the manuscript accordingly in the correct direction. Overall, quality of the paper has been improved. However, there are still some issues for further consideration:
1> The title, in its present form, is appeared as a sentence, which needs to be revised as using sentence as title is rare and unusual.
   • We introduce a new title which is more common to this measurments the title will be: “Experimental study of the inverse diffusion flame using high repetition rate OH /acetone PLIF and PIV” instead of “High-speed imaging of OH and acetone PLIF reveals the structure of methane inverse diffusion flames”.

2> There are still some grammatical and typo errors in the manuscript.
   • The paper is reviewed by many English native languish collegues and all the grammatical and typo errors are corrected.

3> The introduction reviews the studies of IDF, but no measurement using similar laser technique in the literature was quoted and reviewed. If no such previous measurement exists for IDF, the author may refer to other flame types in which laser technique has been used to observe the flame structure. Papers suggested for reference:
   • We added paragraph to highlight the techniques used in the normal diffusion flames which are related to the techniques used in the current work and we included these two references as they have a technique for a temperature measurements in flames. We updated the references list according to this modifications.
Highlights

- The fate of spatial flame structures of methane inverse diffusion flame was monitored and investigated.
- The flame structure is composed of a fuel entrainment region and an intense combustion region.
- Three types of events were common to all flames: breaks, closures and growing kernels.
- In upstream regions of the flames, the breaks were counterbalanced by flame closures.
Experimental study of the inverse diffusion flame using high repetition rate OH \textit{acetone} PLIF and PIV

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Abstract

Most previous work on inverse diffusion flames (IDFs) has focused on laminar IDF emissions and the soot formation characteristics. Here, we investigate the characteristics and structure of methane IDFs using high speed planar laser-induced fluorescence (PLIF) images of OH, particle image velocimetry (PIV), and acetone PLIF imaging for non-reacting cases. First, the flame appearance was investigated with fixed methane loading (mass flux) but with varying airflow rates, yielding a central air jet Reynolds number (Re) of 1,000 to 6,000 (when blow-off occurs). Next, it was investigated a fixed central air jet Re of 4500, but with varied methane mass flux such that the global equivalence ratio spanned 0.5 to 4. It was observed that at Re smaller than 2000, the inner air jet promotes the establishment of an inverse diffusion flame surrounded by a normal diffusion flame. However, when the Re was increased to 2500, two distinct zones became apparent in the flame, a lower entrainment zone and an upper mixing and combustion zone. 10 KHz OH-PLIF images, and 2D PIV allow the identification of the fate and spatial flame structure. Many flame features were identified and further analyzed using simple but effective image processing methods, where three types of structure in all the flames investigated here: flame holes or breaks; closures; and growing kernels. Insights about the rate of evolution of these features, the dynamics of local extinction, and the sequence of events that lead to re-ignition are reported here. In the lower entrainment zone, the occurrence of the flame
break events is counterbalanced by closure events, and the edge propagation appears to control the rate at which the flame holes and closures propagate. The rate of propagation of holes was found to be statistically faster than the rate of closure. As the flames approach blow-off, flame kernels become the main mechanism for flame re-ignition further downstream. The simultaneous OH-PLIF/Stereo PIV measurements indicate that the individual breaks events could be correlated to local vortical flow structure and strain rates fields. The detailed measurements provide a more complete understanding of IDF flame characteristics and structure than was previously possible.

1. Introduction

Inverse diffusion flames (IDFs) are a particular type of diffusion flame with an inner air jet surrounded by an outer fuel jet, which is then surrounded by either ambient air (in practical settings) or an inert gas (in some laboratory settings). Inverse diffusion flames have characteristics of both ordinary diffusion and premixed flames and, hence, can exploit the advantages of the low soot emissions inherent in premixed flames and the flame stability, ease of fuel delivery, and operational safety inherent in diffusion flames. Because of these characteristics, the feasibility of using IDFs in industrial and domestic heating processes has motivated a number of experimental and numerical studies [1-17].

Wu and Essenhigh’s [1] group was one of the first to investigate IDFs experimentally. Six different types of IDFs were mapped out and defined according to the appearance and stability of the flames. The characteristics and structure of natural gas IDFs were investigated by Sobiesiak and Wenzell [2]. They concluded that the inner air jet and outer fuel jets strongly impact the normalized flame length. The low NOx emission characteristics of IDFs were reported [3, 4] in relation to air staged burners. Their main conclusion was that the maximum
emission index of NO\textsubscript{x} occurred at an equivalence ratio of one [3], and peaked just above the outer edge of the flame tip [4]. Sze \textit{et al.} [5] investigate the appearance, temperature distribution, and NO\textsubscript{x} emission index of two inverse diffusion flames, one with circumferentially arranged ports (CAPs) and the other with co-axial jets (CoA). They proposed that the CAP, compared with CoA, was able to operate stably at a lower Reynolds number. A study was conducted on the effects of the air nozzle length on pollutant emission from an IDF burner with multiple fuel jets [6]. It was concluded that the short nozzle yielded a flame with a more intense mixing, resulting in more complete combustion than did the long nozzle. Recently, for CAPs arrangement, the effect of hydrogen percentage to LPG and air jet Reynolds number on the lean stability limit were investigated [7].

A number of investigations on IDF sooting tendencies have also been performed [8-9], where the soot forms on the outside of the flame [8, 10]. Soot collected from IDFs is observed to be tar-like with a high hydrogen content [11, 12]. It is similar to the soot collected from under ventilated normal diffusion flames [13]. Recently, the flame structure of laminar inverse diffusion flames was studied by Mikofski \textit{et al.} [14] to gain insight into soot formation and growth during under ventilated combustion.

Regarding the flow field in IDFs, some authors have presumed that the velocity field of IDFs is similar to that of a normal diffusion flame [15]. A schematic of the laminar flow field of IDF flames was presented by Belivins \textit{et al.} [13]. In this schematic, the flow field indicated inward flow near the base of the flame, which was followed by slight expansion (with small radial outflow) near the flame tip. Recently, the effect of the fuel-to-air velocity ratio on the IDF\textsubscript{s} structure was described [16-17] using OH/PIV measurements but with an annular separating distance between the air and fuel jets. In these studies, there was a separating
distance between the annular fuel jet and the central air jet. This separating distance was found to play a significant role in the flame structure and flame stability. On the other hand, the normal diffusion flame has been investigated extensively using different measuring techniques. High repetition rate OH-PLIF in a turbulent non-premixed methane flames was used to resolve the turbulent flow phenomena such as vortex formation and air entrainment [18], and to investigate the extinction and re-ignition in non-premixed flames with different levels of oxygenation [19]. However, more insight into the flame-hole re-ignition mechanisms in turbulent non-premixed flames was gained through the use of simultaneous high speed PIV/OH-PLIF give [20, 21]. The temperature fields in butane-air impinging flames and slot laminar jet flame were explored and investigated using reference-beam interferometry [22, 23], where they showed that the two dimensional temperature field could be calculated through the application of the inverse Abel transformation.

In this work, we investigate the characteristics and structure of methane IDF's. Two sets of experiments were conducted: the first set studied the effect of the Re of the air jet on the flame structure with the fueling rate fixed; the second set investigated the effect of the fuel flow rate on the flame structure at a fixed air flow rate. The flame structure was investigated using high-speed OH-PLIF images of flames at 10 KHz collected along the flame, acetone vapor PLIF measurements for tracing the fuel stream under non-reaction flame conditions, as well as 2D PIV flow-field measurements. To explore the physical behind the local flame extinction and re-ignition, simultaneous OH-PLIF/Stereo PIV measurement was conducted on some selected flames.

2. Experimental setup and diagnostics

2.1. Burner and flames
The burner geometry used in this study is shown in Fig. 1a. It was constructed using two vertical concentric stainless steel tubes. The inner tube carried air and had an inside diameter of 5.76 mm, while the inner diameter of the outer tube was 10.8 mm. The annular opening between the tubes delivered methane. The burner was surrounded by a Pyrex shield with a side opening to admit laser light and allow for imaging. The shield was used to mitigate the flame instabilities that could be caused by room air currents. First, the flame appearance was investigated with fixed methane loading (mass flux) but with varying airflow rates, yielding a central air jet Reynolds number (Re) of 1,000 to 6,000 (when blow-off occurs). Next, it was investigated a fixed central air jet Re of 4500, but with varied methane mass flux such that the global equivalence ratio spanned 0.5 to 4. The flame conditions, the global equivalence ratio and their designations are given in Table 1. Among the flames appearance conditions, six were selected for detailed reaction zone and mixing imaging via 10 kHz OH-PLIF and acetone PLIF respectively, and five of these were further investigated using velocity field (via 500 Hz PIV). Three of the flames with a constant Re of 4500 and varying fueling rates were selected for OH/acetone PLIF imaging and four were selected for PIV.

2.2. Planar laser-induced fluorescence (PLIF) of OH and acetone vapor

Planar laser induced fluorescence (PLIF) is carried out on OH radicals to track the spatial and temporal features of a flame’s structure at a 10 KHz repetition rate. The experimental configuration is illustrated in Fig. 1b. The excitation of OH radicals is carried out using a frequency-doubled high-speed dye laser (Sirah, Cerdo-Dye) pumped with a frequency-doubled, diode-pumped solid state INNOSLAB laser (Edgewave IS16II-E). The dye laser produced a fundamental beam at 566 nm with Rhodamine 6G. This light was then frequency doubled using a BBO crystal to produce ultraviolet beam at 283 nm. The laser wavelength was tuned to 283.01
nm, to pump the $Q_1(6)$ transition of the $(1, 0)$ band of the $A^2\Sigma \leftarrow X^2\Pi (1, 0)$ system of OH. The laser produced 2.4 W (time averaged) power at 283 nm at 10 KHz. The laser beam was expanded through two cylindrical lens, creating collimated sheets approximately 100 mm in height. It was then focused to a waist using a third cylindrical lens with a focal length of 600 mm. The laser sheet was approximately 130 µm at the center of the burner.

Fuel distribution in the flow field (under non-reacting flame conditions) was characterized by introducing acetone as a tracer to the fuel stream; the acetone was seeded by bubbling the methane through liquid acetone. To avoid fluctuations in the acetone seeding density due to temperature fluctuations, the container of acetone was immersed in a thermostatic bath (constant temperature of 350 K). In this work, the acetone was excited at the same wavelength as OH radical (at 283 nm). A seeding level lower than 10 % by mass of the acetone in methane was used. This concentration was high enough to ensure a reasonable level of fluorescence yet low enough to observe the same flame dynamics as in flames without acetone.

A CMOS ICCD camera was used to capture either OH or acetone fluorescence. The camera was combined with appropriate filters to collect the fluorescence signals, for the OH-PLIF imaging, a good discrimination of the OH excitation frequency was achieved using a transmission band-pass interference filter > 80% at 310 nm, (custom fabrication- Laser-Components GmbH). Standard filters WG320 (high-pass at 320 nm) and SWP606 (low-pass at 500 nm) were used to acquire acetone vapor fluorescence photons. Each OH/acetone PLIF images was processed to remove (a) laser sheet inhomogeneity and (b) noise in the background from shot-to-shot laser fluctuations. The raw fluorescence image was corrected by taking into account the actual profile of the laser beam and the background signal. The sheet’s profile correction was determined using an ensemble average of 10,000 individual images of the laser
sheet passing through a cuvette filled with acetone placed in the center of the field of view. An average was then extracted from these raw images and used to correct the fluorescence signal.

2.3. PIV set up

As shown in Fig. 1b, a PIV system was used to acquire 2D velocity measurements in a plane at a repetition rate of 500 Hz, with the same field of view as the PLIF system. The PIV system consisted of a dual-cavity, diode-pumped, solid state Nd: YLF laser (LDY 300 Series) and a single CMOS camera (La Vision, image pro HS 4M); the laser was capable of delivering 35 W per head at 10 KHz, with a 9 ns pulse duration. With three cylindrical lenses, the laser pulses were formed into a sheet with a 0.5 mm waist along the burner’s centerline. Both the air and fuel jets were seeded with titanium dioxide (TiO\textsubscript{2}) particles with nominal diameters of 0.5 µm. Mie-scattered light from the seeding particle-laden flow was collected using a 105 mm, f/4 objective lens (Nikon UV Micro-Nikkor) equipped with a 527 nm band pass filter. An adaptive multi-pass vector evaluation procedure was used, with interrogation areas ranging from 128 pixels to 24 pixels. This resulted in a spatial resolution and vector spacing of approximately 0.5 mm and 0.25 mm, respectively.

3. Results and discussion

3.1. Flame appearance

The effect of the air flow rate, in terms of the exit air Re, on the flame’s appearance (chemiluminescence and natural soot incandescence) for a given fuel flow rate of 1.77 slpm is shown in Fig. 2a. In conventional diffusion flames, the fuel burns with the air entrained from its surroundings and the flame is highly luminous due to soot incandescence. The introduction of a central air jet leads to the establishment of an inverse diffusion flame inside the conventional
diffusion flame. At low air jet velocities (1000 ≤ Re < 2000), both flames are attached to the nozzle tip, and the flames exhibit a similar appearance to a pure diffusion flame. It can be observed that increasing the air jet velocity results in lifting of the IDF and shortening of the outside flame. The appearance of a dual flame structure (a bluish reaction zone confined in a diffusion combustion zone) occurs in flames at Re = 2000. Increasing the air jet velocity (Re ≥ 2500) causes blow-off of the inner flame and establishment of an intense blue, bell-shaped structure inside the flame. Further increases in the air jet velocity (Re ≥ 4500) lead to an opening of the flame tip and eventually lead to complete flame blow-off at Re = 6000.

Figure 2b presents examples of single-shot acetone vapor PLIF images of some of the previously discussed flames with the air jet Re between 3000 and 5500. These acetone-PLIF images as well as the flame appearance provide new information on IDFs. The fuel is entrained immediately into the air jet at the burner exit when Re ≥ 3000, leading to a flame consisting of two regions: a lower zone where the fuel is entrained toward the central air jet and an upper zone where the fuel mixes with air, leading to intense combustion. These two regions are separated by a flame neck. This flame neck is surrounded by a soot ring at Re = 2500, and this sooty ring gradually disappears with increasing Re number (Fig. 2a). The appearance of this sooty ring indicates non-premixed combustion of the fuel jet before entrainment into the central air jet. Increasing Re from 3000 to 4000 leads to increasing fuel entrainment in the lower zone of the flame, as indicated by a decrease in the length of the fuel entrainment region. However, further increases in the Re number impose a higher strain rate at the end of the entrainment region, which leads to less opportunity for the fuel to mix with the central air jet. This is clearly observed from the penetration of the central jet into the fuel entrainment region as the flames seen from top view (Fig. 2a, at Re ≥ 4500), which yields a higher mixing rate further
downstream of the jet but still within the shear layer, as indicated by acetone measurements (Fig 2b).

The effect of the fuel flow rate on the flame’s appearance was investigated with a constant airflow rate yielding a Re = 4500 is shown in Fig. 2c. The overall equivalence ratios for these flames range from 0.5 to 4. As seen in this figure, the flames are mostly blue, with a ring of soot appearing when the equivalence ratio exceeds Φ = 2.5 (at Φ = 2.0, the soot ring is just visible) and becoming more obvious with increased fuel flux. For flames with an equivalence ratio of Φ < 1, the air jet is able to penetrate through the central region of the flame, leading to a hollow flame structure. Flames with 1 ≤ Φ ≤ 1.5 are mostly blue with a thick fluctuating reaction zone that can be clearly observed at the flame’s downstream central region with the early annular laminar flame confined to the central jet. This suggests that the combustion mode is primarily premixed in this central region, which is remarkable for Φ = 1.5 flame. Flames with an overall equivalence ratio of Φ > 1.5 exhibit a more intense blue color along the flame’s boundaries, implying that most of the combustion occurs in the shear layer between the air and fuel jets. This is made more clear by considering the single-shot, acetone-PLIF images shown in Fig. 2d of non-reacting jets at Re = 4500 with three overall equivalence ratios (Φ = 0.89, 1.5 and 2).

With increased fuel flux, the effect of the fuel entrainment gradually diminishes in the early entrainment region. Image at Φ = 2 indicates that the annular fuel jet basically co-flows with the central air jet over a long distance of the lower part of the flame. Additionally, the flame neck, obvious at Φ = 0.89, disappears in the richer flame. This indicates that with the increase of the axial momentum associated with the annular fuel jet, the effect of the entrainment region in the lower part of the flame is successively reduced.

3.2. Time sequence of OH-PLIF images
OH-PLIF imaging was used to determine the reaction zones [24-25] in the flames studied here. Time sequences of instantaneous OH-PLIF images for flames with different air-jet Re and a constant fuel flow rate are shown in Figs. 3-5. The time interval between any two consecutive images in a given row is 0.1 ms and the three-row composite images of OH-PLIF cover a physical downstream distance of 86 mm along the flame centerline. Time sequences of OH-PLIF images for flames with Re = 3000 and 3500 (F_{3000,1.34} and F_{3500,1.15}; the flame designation is signified by F with a subscript Re followed by a hyphen and the global equivalence ratio) are shown in Fig. 3. The OH images indicate the presence of a round, short flame attached to the nozzle tip. This anchored flame consumes a small fraction of the available fuel with the remainder abruptly convected towards the inner air jet and subsequently entrained by the jet flow. This leads to thin OH filaments (laminar flame) in the upstream regions of the flame near the burner exit and confines the fuel entrainment region, as shown in flame F_{3000,1.34} (Fig. 3a).

Eventually, the filaments grow toward the central region of the flame. However, in flame F_{3500,1.15} (Fig. 3b), the annular laminar flame covers only a short portion of the flame. In these flames, the OH zones gradually gain thickness and intensity as the downstream distance increases and these zones eventually grow into the central region of the flame. In addition, the more corrugated structure of the OH zones and the broadening in OH distribution are observed in this flame, suggesting a larger partial premixed region that develops into a well-mixed reaction zone further downstream at this higher Re. This may be due to the higher fuel entrainment rate and hence more intense fuel/air mixing, as indicated by the shorter fuel entrainment region observed in the acetone-PLIF images. The greater prevalence of OH zones in the center region of the higher Re flames compared with the lower Re flames in the upstream regions is consistent with an increased rate of fuel/air mixing at the higher Re.
Najm et al. [26] showed that the disruptions in an otherwise continuous OH profile agree well with local flame extinction. In most of the flames studied here, frequent local extinctions or breaks are observed (marked with red arrows in the figures). These breaks were observed to grow and lead to an enlarged gap as they propagated downstream. After the onset of extinction, as shown in Fig. 3a, the edge flames along the perimeter act as an extinction front and the breaks grow with time. The growth of these breaks does not, however, continue indefinitely; at some point, the locally extinguished regions reignite. These local extinction events are counterbalanced by the occurrence of closures, which reconnect the flame sheet and maintain stable combustion particularly in the upstream regions of the flames. Selected sequences of individual closure events are marked by the red-dashed boxes in Fig. 3b. Small flame kernels are also observed in the flames. These flame kernels tend to grow and propagate as they travel downstream where more combustible conditions are met and growth of flame kernels can be sustained, as shown in Fig. 3, where this mode of growth is illustrated by the sequences of red circles.

The OH-PLIF time sequences of flames $F_{4000-1}$ and $F_{4500-0.89}$ are shown in Fig. 4. For flames with $Re > 3500$, pockets of cold fluid are more frequently observed at flame central region. In these flames small fraction of the OH zone is observed in the central region of the flame, with the majority of the OH zone residing on the boundaries of the flame, as seen in Fig. 4b. This illustrates the penetration of the cold mixture into the center of the flame, consistent with the flame luminosities shown in Fig. 2a. This leads to an increased occurrence of disconnected OH zones and demonstrated by a comparison of Fig. 3 with Fig 4. These intermittent regions of OH tend to reconnect into a more uniform flame in the region of the second row in flame $F_{4000-1}$ (between 25 and 50 mm from the nozzle tip), or further downstream in flame $F_{4500-0.89}$. This
reconnection mode is different from the closure mode due to edge propagation as discussed earlier (occurring further upstream). These kernels initiate from the inner corrugated surfaces of the annular OH layer and grow as they travel into regions far from the flame’s centerline, where local strain rates are lower and the surrounding conditions more favorable, reconnect the breaks events further downstream locations. Samples of these growing kernels are given in the second row of images in Fig. 4b. The occurrence of flame kernels is more frequently observed in flames approaching global blow-off with higher Re. However, at flame upstream, flame hole closure events are observed upstream, as shown in the first row of images in Fig. 4b, still due to flame segment propagation.

The instantaneous structures of OH zones in flames with Re ≥ 5,000 are quite different from those with comparatively lower Reynolds numbers, as seen in Fig 5. In these high Re flames, the reaction zone is highly contoured, with significant fragmentation occurring. In general, the structure of the OH zones of flames with high Re and approaching the flame blow-off limit is characterized by a large fraction of disconnected fragments, and these fragments do not exhibit the combination of premixing and diffusion in the OH-zone structures as observed in the lower Re flames. In addition, the flame kernels seem to serve as re-ignition sources in the flame closure events of flames at higher Re numbers.

The effect of the fueling rate on the OH-zone structures of flames with an air jet Re of 4,500 and global equivalent ratios of 0.89, 1.5 and 2 are shown in Figs. 4b, 6a and 6b, respectively. It is apparent that both the equivalence ratio and the air-to-fuel jet momentum ratio strongly impact the structures of the OH zones. As mentioned previously, flame F_{4500-0.89} generally exhibited a corrugated and thick OH-zone structure residing on the boundaries of the flame, resulting from the penetration of the central jet into the inner region of the flame. In flame F_{4500-
thin OH zones are observed in the upstream regions of the flames near the burner exit and extend nearly two-thirds of the flame length. This flame is accompanied by frequent occurrences of flame breaks. Further downstream, the OH zone gradually thickens and eventually prevails across the entire central region of the flame, as shown in Fig. 6a. These well-distributed OH zones are consistent with the OH-zone structures seen in premixed flames, suggesting a combined air-fuel mixing mode (premixed-like and pure diffusion flame-like modes) for flames with $\Phi = 1.5$. These results agree with the acetone-PLIF images, where the higher fuel concentrations are confined to the early air regions leading to the disappearance of the flame necking. This indicates less mixing in the upstream region of the flame.

In flame $F_{4500-2}$, comparatively thinner OH zones are observed on the boundaries spanning the entire imaging region, as seen in Fig. 6b. This OH-zone distribution is consistent with OH distributions observed in normal diffusion flames. In this case, the acetone-PLIF image indicates the disappearance of the fuel entrainment region, which leads to less entrainment in the lower part of the flame. On the other hand, the increase in the equivalence ratio (by increasing the fueling rate) leads to an increase in the fuel jet momentum and a corresponding decrease in the air-to-fuel jet momentum ratio, which in turn reduces the shear between the air and the fuel jet. The decreased shear is expected to reduce the mixing between the fuel and air, leading to the observed thin OH zones.

3.3. Propagation rates of flame closures and flame breaks

The propagation rates of flame closures and breaks were calculated using the following approach. The principle is to use a 3 x 3 Gaussian smoothing filter on the OH images that are binarized by a specific threshold and then examine different threshold values to ensure that the chosen threshold has no significant impact on the determination of the size of the flame breaks.
This approach allows the length/size of the flame hole to be determined and, since the frame rate between any two consecutive images is known, the propagation rate of the flame closures and flame breaks can be determined (positive rates for closures; negative rates for breaks). Selected examples of an OH-PLIF image time sequence are used to determine the rates of flame breaks and flame closures and are shown in Fig. 7. The corresponding binarized OH-PLIF images are shown in the bottom of the plots, and the growth and propagation rates for flame breaks and closures are shown to the right of the plots.

There are many factors that may affect the correct identification of these flame break or closure events and their corresponding propagation rates. In the current study, sequences of a flame break are only considered if the flame edge extinction is vertically aligned. This therefore excludes any horizontal shift of the flame, where the flame edges are more likely to be initiated from the same flame sheet. Also, any out-of-plane motion of the flame structures could result in erroneous interpretations of the OH images. To mitigate the impact of out-of-plane motion, two conditions must be met before the absence of an OH signal is deemed a flame break. First, if the structure is coming from out-of-plane motion, it is more likely to be stagnant on the plane of measurement. By comparing the OH images containing the potential holes with the corresponding measured average axial velocity at that location, and knowing that the time between OH images is 0.1 ms, the minimum threshold by which the flame structure can be shifted due to advection can be calculated. Since the velocity measurements are made at a 500 Hz repetition rate, relatively slow in comparison with 10 KHz OH system, the estimation of the threshold distance is based on an average flow field measurement. Figure 8 illustrates the mean velocity vector over the entire length of flame F_{3000-1.34} with instantaneous OH images containing a flame hole. The mean axial velocity at the flame break location is 5.5 m/s, which
corresponds to 0.55 mm average motion between two consecutive OH images. This distance is then taken as a minimum threshold by which the flame structure needs to move to be considered as a sequence for hole or closure calculation. The hole propagation and/or closure rate should be approximately constant over a short period; if the flame structure is dominated by out-of-plane motion, it will have a rate that arbitrarily changes over time. Consequently, only sequences of images that have either monotonically increasing or decreasing gap widths are considered as true hole and closure events, respectively. Secondly, the evaluation of flame break or closure events is restricted to regions close to the burner exit, where the flow is laminarized in the reaction zone due to the heat release and corresponding increase of viscosity. Here, the OH layers are thin and restricted to the reaction zones. The validity of OH as a reaction zone marker in these lower regions has been confirmed in previous studies [27].

A local deviation from thermal equilibrium in which the rate of heat transport away from the flame surface can exceed the rate heat release from the chemical reactions could lead to the local extinction of the flame and the formation of the flame breaks. This deviation is due to the turbulence flame interaction. The more frequent events of breaks and the large size of breaks, negatively affect the flame stability, combustion efficiency and pollutant emissions from practical points of view.

To investigate the possible physical phenomena taking place in the turbulence/chemistry interactions, which leads to the observed breaks and closure events, simultaneous Mie scattering (Stereo PIV)/ OH-PLIF imaging was conducted on flame F_{4500-0.89}. Four components of the strain rate tensor was calculated from the 2D plane velocity components. The strain rate tensor was then transferred to principle coordinates to extract the two principle strain rates. The maximum principle strain rate components corresponds to extensive strain, and the minimum
principle component corresponds to compressive strain. The velocity vector measurements showed that the third velocity component (out of plane) was not significant, especially near the burner exit. Consequently, the principal strain rates computed from 2D measurement data are valid near the burner exit. As shown in Fig. 8a-b, two examples of local breaks events, where the disruptions in otherwise continuous OH fronts correlate well with the local flame front’s extinctions. Measurements were performed close to the burner exit where the flow is laminarized in the reaction zones. In this domain, the OH layers are thin and restricted to the reaction zones. As indicated, the break events seem to be caused by a large flow vortex breaking through the OH fronts. This flow vortex impinges on the flame front and the extinguished flame tips are seen to correspond with regions of very high extensive strains (see the zoomed-in window to the top right of each plot). At extinction, the strain rates are high on the air side of the OH layer, and there is a strong velocity component directed towards the flame front. This leads to an increased rate of heat loss associated with increasing scalar dissipation rates. On the other, the flow field associated with closure events or continuous OH layer is seen to be laminarized by the higher viscosity burned gases, which provides a shield, preventing the flow vortices from penetrating through the flame fronts and correspond to low strain rates (see the right part of each plot). The closure event seems to be mainly due to the edge flame re-ignition mode. These findings agree with some observations on the extinction of normal diffusion flames, where in the normal diffusion beside extinction due to the high strain rate associated with large flow vortex impinging on the flame front, sometimes the flame is extinguished at much lower strain rates than the instantaneous peak strains. It is seems that the present flames are subjected to such high strains for only short times and becomes more frequent as the flames approach the global extinction.
As suggested by Steinberg et al. [20], flame closure events could occur due to the propagation of flame segments, the advection of flow turbulence, or both. In the current study, the effect of flow advection on the rate of flame closures could be estimated from the average velocity field (as indicated for flame F3000-1.34, Fig. 9). This yields a difference of 0.4 m/s between the propagation velocities upstream and downstream of the flame tips, or about 7.4% of the average propagation rate of flame closures (≈ 5.4 m/s), suggesting that the effect of flow advection on flame closures is small and the propagation of flame segments has a relatively strong impact on flame closures.

Histograms of the propagation rates of flame closures and flame holes are shown in Fig. 10. for flames F3000-1.34, F4500-0.89, and F4500-2 (top row for closure and bottom for breaks). These results were constructed for events occurring on the right flame edge in the image and at a downstream distance of 28 mm from the burner exit. As shown in Fig. 10, flames with different Re and Φ exhibit some similarity in the distribution of the flame closure propagation rate, but with different mean values of this rate. For example, for the flame with Re = 3000 (F3000-1.34), the overall mean propagation rate of flame closures is 5.4 m/s and the highest occurrence of flame closures is at a velocity of nearly 2.5 m/s. In addition, a closure propagation rate larger than 6.5 m/s is found to occur less than 27% of the time. An increase in Re to 4500 (F4500-0.89) leads to a slight decrease in the overall mean propagation rate of flame closures, to 5.1 m/s, while the occurrence of the closure propagation rate larger than 6.5 m/s also decreases to 11.4%. The overall mean closure propagation rate further decreases to 4.1 m/s as the equivalence increases to 2 for the flame F4500-2.0, as shown in Fig. 10c. A relatively higher occurrence of low closure propagation rates is also observed for flames with Φ = 2, possibly due to a decreased
shear between the fuel and the air, leading to a decrease in the rate of entrainment and mixing, as discussed previously.

For flames with a Re = 3000 (F_{3000-1.34}), an overall mean hole propagation rate of -7.9 m/s and a peak occurrence of -2.0 m/s are observed, as shown in Fig. 10d. The mean hole propagation rate is relatively higher than that of the closure propagation rate (≈ 5.4 m/s) and the propagation rate of flame holes generally covers a wider range than that of flame closures. A hole propagation rate greater than 6.5 m/s occurs about 45% of the time, compared with less than 27% of the time for flame closures. A mean hole propagation rate higher than that of the closure rate is also observed for flames with increased Re (see Fig. 10e) and for flames with higher equivalence ratios (see Fig. 10f). As the Re is increased to 4500, (F_{4500-0.89}), the occurrence of low hole propagation rates decreases, accompanied by an increase in the mean value of the hole propagation rate to -8.2 m/s. Also, flames at Φ = 2 (F_{4500-2.0}), show a higher probability of hole propagation rates, with an average overall hole propagation rate of about -5.4 m/s (Fig. 10f). These results indicate that with increasing Re, the difference between the hole propagation rate and the closure propagation rate increases. This means as the flame approach the global flame extinction conditions, the difference between the hole and closure propagation rates is increased, and hence the importance of flame hole size investigation.

3.4. Center line velocity profiles

The axial distribution of the mean centerline velocity (V_{cen}) normalized by the maximum centerline velocity (V_{max}) and the corresponding velocity fluctuations (V_{rms}) with different Re (F_{3000-1.34}, F_{3500-1.15}, F_{4000-1.0}, F_{4500-0.89}, and F_{5500-0.8} under a constant fueling rate, Fig. 11a, Fig. 11c) and for flames at Re = 4500 with different global equivalence ratios (F_{4500-0.89}, F_{4500-1.5}, F_{4500-2.0}, and F_{4500-3.0} Fig. 11b, Fig. 11d), are shown in Fig. 11.
As shown in Fig. 11a, the centerline velocities for these flames, are remarkably different. At relatively low Re (F_{3000-1.34}, F_{3500-1.15}), the centerline velocity displays a nearly constant value and is followed by a slight decrease at an axial location between 30 to 40 mm from the burner exit. Further downstream, the centerline velocity steadily increases, reaching a peak around 50 mm downstream. For flames F_{4500-0.89} and F_{5500-0.73}, the centerline velocity profiles are nearly constant for a short axial distance from the burner exit followed by a slight but steady increase. The maximum velocity is located near the same position observed for the lower Re flames, but with an early decrease in the mean velocity in flame F_{4500-0.89}. A further increase of Re to 5500 (F_{5500-0.73}) leads to a more rapid centerline decay. For these high Re number flames, the central air jet was able to penetrate through the flame, resulting in the dramatically lower flame temperatures.

With reference to the OH-PLIF images, it is expected that the velocity field will be affected by the heat release rate and related to the OH profile. Flames with relatively low Re (Fig. 3a, F_{3000-1.34}) have a higher OH intensity along the flame centerline than do those of higher Re (Fig. 5b, F_{5500-0.73}) at Y > 50 mm. This could explain the later decrease in the centerline velocity in the lower Re flames, with the higher intensity (at lower Re) OH signal indicating an increase in the temperature and the expansion of gases as they are advected from the unburnt side to the burnt side. The centerline velocity fluctuations for three lower Re flames show relatively small fluctuations along the flame length with a slight decrease in the magnitude of fluctuations between Y = 50 to 60 mm, followed by a relatively slight increase at the flame tip, as shown in Fig. 11c. However, for flames F_{4500-0.89} and F_{5500-0.73}, where the mean centerline velocities show a much more pronounced decrease but at different axial locations, the region of velocity decline is associated with an obvious increase in the centerline velocity fluctuations, with higher levels.
of fluctuations at Re = 5500. The damping of fluctuations over the entire centerline of lower Re
flames, and over nearly 50% of the flame length for Re = 4500 and 5500, is in agreement with
the numerical studies of jet diffusion flames by Yamashita et al. [28]. These researchers
concluded that the increased viscosity in the flame jet decreases the amplitude of the fluctuating
flame surface through the suppression of the velocity fluctuations.

The effect of the fueling rate can be understood from close inspection of the mean centerline
velocity. Figure 11b shows the normalized centerline velocities for different equivalence ratios.
The centerline velocity of flame F4500-0.89 increases slightly to a constant value over a wide span
of the flame length. However, after increasing Φ to 1.5 (F4500-1.5), a slight decrease in the
centerline is observed, followed by a nearly constant centerline velocity region and ending with
a slight increase in velocity leading to its maximum velocity at flame tip. These results are in
agreement with the previous discussion of the OH images shown in Figs. 4b and 6a, where
increasing Φ to 1.5 affects the degree of partial premixing between the upstream fuel and air.
This leads to an increase in the flame temperature, sustains the flame centerline velocity over
the flame tip, and consequently leads to a suppression of the velocity fluctuations.

Increasing Φ to 2.0 delays the fuel-air mixing, and leads to a significant change in the OH
distribution as shown in Fig. 6b, relative to flame F4500-1.5 (Fig. 6a). This change in OH
distribution, with no OH signal detected along the flame centerline, leads to a continuous
decline in the centerline velocity as indicated in Fig. 11b. This decline in velocity is
accompanied by a nearly constant low level of velocity fluctuation, with a very slight velocity
increase at the flame tip (Fig. 11d). A further increase in the equivalence ratio to Φ = 3.0 leads
to the same features as observed for Φ =2.0, but with a steeper decrease in the centerline
velocity (Fig. 11b). This suggests that the degree of entrainment and hence for the flames with
Φ from 1.5 to 3.0 are inversely proportional to the equivalence ratio, which means that for these fuel flow rates, the fuel-air mixing is reduced and delayed with the obvious suppression in the velocity fluctuations.

4. Conclusions

Axisymmetric inverse diffusion flames (IDFs), in which the fuel jet surrounds an inner air jet, have many practical applications. High-speed images from OH-PLIF, 2D PIV, and acetone PLIF were used to investigate the structure of methane IDFs. First, the flame appearance was investigated with fixed methane loading (mass flux) but with varying airflow rates, yielding a central air jet Reynolds number (Re) of 1,000 to 6000 (when blow-off occurs). Next, it was investigated a fixed central air jet Re of 4500, but with varied methane mass flux such that the global equivalence ratio spanned 0.5 to 4. At low Re number, an inner IDF is formed inside the normal diffusion flame and a unique double flame structure is observed, where the reaction zone is enveloped by an outer diffusion flame. With further increase in Re number both flame appearance and acetone PLIF images demonstrated that the flames consist of two regions, a lower entrainment region and a mixing and combustion upper region. In the lower entrainment zone, the IDF exhibits a varying degree of partial premixing at upstream locations, depending on the Re and equivalence ratio. An area of increased turbulent mixing along the shear layer of the central jet then develops into a well-mixed reaction zone downstream of the flame. At lower global equivalence ratios, fuel entrainment enhances fuel air mixing in this lower entrainment zone, but this effect diminishes with as the equivalence ratio for increases in the high Re cases. From these detailed descriptions, it is possible to conclude that the degree of entrainment of the fuel jet by the air determines the extent to which the fuel is burned in the diffusion mode or
premixed mode. This degree of entrainment depends on the relative momentum of the air and fuel jets.

Although qualitative in nature, the OH-PLIF images possess sufficient spatial and temporal resolution to allow quantification of the evolution of extinction and re-ignition events in these flames. Three types of events common to the IDF flames studied here. The first type of event is breaks that mark the initiation and further widening of gaps in an otherwise continuous OH-PLIF profile. These breaks correspond to the well-known local extinction events in a wide range of ordinary non-premixed flames approaching extinction. The second event is closure events, which refer to the gradual reconnection of the extinguished edges. Particularly at locations near the fuel tube, the occurrence of the flame break events is counterbalanced by closure events. A correlation between break events and both the local strain rates and vortical flow structures is existed. On the other hand, laminarization in the reaction zone provides an effective shield preventing flow vortices from penetrating the flame front. The results show that the rates of propagation of a flame hole is typically faster than that of a closure. With increasing Re, the difference between hole propagation and closure propagation rates increases, which increases the probability of flame blow-off. The third event is the growth of kernels, which are initially small kernels of OH that grow with time. The occurrence of these flame kernels increases as the flames approach global extinction within the flame sheet where there is significant broadening of the OH layer. These kernels are elongated and develop into fully connected flames further downstream and became the main mechanism in flame closure events in regions downstream of the flame and in flames with higher Re, which approaching global blow-off.

Acknowledgments
The work described in this paper was supported by the Clean Combustion Research Center (CCRC), King Abdullah University for Science and Technology (KAUST). The burner was designed at Faculty of Engineering-Materia, Helwan University.

References


Table 1: Operating conditions for inverse diffusion flames

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<thead>
<tr>
<th>Re</th>
<th>Φ</th>
<th>Flame appearance</th>
<th>Conditions with a fixed fuel flow rate</th>
<th>Acetone-PLIF</th>
<th>PIV</th>
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<th>PIV</th>
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<td>F4500-4</td>
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* Flame appearance or measurements are presented for these conditions.
Fig. 1. (a) Schematic diagram of the IDF burner; (b) Schematic diagram of the experimental setup for PIV and OH-PLIF set up.
Fig. 2. (a) The effect of air flow rate on the appearance of IDF at a fixed fueling rate of 1.77 slpm on flame appearance; (b) Single shot acetone PLIF images under non-reacting selected flame conditions (3,000 ≤ Re ≤ 5,500); (c) The effect of overall equivalence ratio on the flame appearance at Re = 4,500; Single shot acetone PLIF images under non-reacting three selected flame conditions (Re = 4,500, Φ = 0.89, 1.5, and 2).
Fig. 4. Selected sequences of OH-PLIF images collected at various axial locations in: (a) flame $F_{4000-1}$; (b) flame $F_{4500-0.89}$. Images in time sequences from left to right, with 0.1 ms between consecutive flames. Different rows of images, the first row covers the axial distance of $0 \leq Y \leq 25$ mm; the second covers the axial distance of $25 \leq Y \leq 50$ mm; and the third row covers the axial location of $50 \leq Y \leq 86$ mm. Images for different axial locations are not in time sequences. Breaks, Closures, and growing kernels are indicated by arrows, squares, and dashed circles, respectively.
Selected sequences of OH-PLIF images collected at various axial locations in flame: (a) $Re = 5000, F_{5000-0.8}$; (b) $Re = 5500, F_{5500-0.73}$

Images in time sequences from left to right, with 0.1 ms between consecutive flames. The first row covers the axial distance of $0 \leq Y \leq 25$ mm; the second covers the axial distance of $25 \leq Y \leq 50$ mm; and the third row covers the axial location of $50 \leq Y \leq 86$ mm. Images for different axial locations are not in time sequences. Closures and growing kernels are indicated by squares and dashed circles, respectively.

Fig. 5. Selected sequences of OH-PLIF images collected at various axial locations in flame: (a) $F_{5000-0.80}$; (b) $F_{5500-0.73}$
Fig. 6. Selected sequences of OH-PLIF images collected at various axial locations in flames: (a) $F_{4500-1.50}$; (b) $F_{4500-2.25}$.

Images in time sequences from left to right, with 0.1 ms between consecutive flames. The first row covers the axial distance of $0 \leq Y \leq 25$ mm; the second covers the axial distance of $25 \leq Y \leq 50$ mm; and the third row covers the axial location of $50 \leq Y \leq 86$ mm. Images for different axial locations are not in time sequences. Breaks and growing kernels are indicated by arrows and dashed circles, respectively.
Fig. 7. (a) Sample images of a break event. Shown are the processed (top row) and binarised (bottom row) images of a flame hole growing in a Re = 3000 flame (F3000-1.34). Plots on the right show the ‘break size’ and calculated ‘flame propagation rate’ versus time; (b) Sample images of a closure event. Shown are the processed (top row) and binarised (bottom row) images of a flame hole closing in a Re = 3500 flame (F3000-1.15). Plots on the right show the ‘closure size’ and calculated ‘flame propagation rate’ versus time.
Fig. 3. Selected sequences of OH-PLIF images collected at various axial locations in: (a) flame $F_{3000-1.34}$; (b) flame $F_{3500-1.15}$. Images in time sequences from left to right, with 0.1 ms between consecutive flames. Different rows of images, the first row covers the axial distance of $0 \leq Y \leq 25$ mm; the second covers the axial distance of $25 \leq Y \leq 50$ mm; and the third row covers the axial location of $50 \leq Y \leq 86$ mm. Images for different axial locations are not in time sequences. Breaks, Closures, and growing kernels are indicated by arrows, squares, and dashed circles, respectively.
Fig. 8. Instantaneous simultaneous Mie scattering and corresponding velocity vectors/OH PLIF imaging. (a-b) Two examples of breaks events (flame $F_{4500-0.89}$), the gray scale is the Mie scattering and is overlaid with OH radicals contours (yellow lines) and velocity vectors (red vectors), the zoomed windows to the right of each plot show the maximum principles strain rates in gray scale and the corresponding OH contours, and velocity vectors.
Fig. 9. (a) Mean velocity vectors, (alternate vectors have been removed for clarity); (b) instantaneous OH-PLIF image that contains a flame hole in $F_{3000-1.34}$, Re = 3000.
Fig. 10. Histogram of the number of occurrences/1500 in relation to rates of closures and holes for flames of Re = 3000, 4500 (F\textsubscript{3000-1.34}, F\textsubscript{4500-0.89}, constant fuel flow rate) and with flame at Re = 4500 - Φ = 2 (F\textsubscript{4500-2}). The closures are defined as positive propagation rates, while the holes are defined as negative propagation rates.
Fig. 11. The mean centerline velocity as a function of the axial distance normalized by the maximum centerline velocity for (a) Re = 3000, 3500, 4000, 4500, 5500 flames (with constant fuel flow rates, $F_{3000}$, $F_{3500}$, $F_{4000}$, $F_{4500}$, $F_{5500}$), (b) for $\Phi = 0.89, 1.5, 2$, and 3 flames (Re is kept constant at 4500, $F_{4500}$). The corresponding centerline velocity fluctuations for the two sets are presented in (c) and (d), respectively.
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