DISPERSION, IGNITION AND COMBUSTION CHARACTERISTICS OF LOW-PRESSURE HYDROGEN-METHANE BLENDS

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ABSTRACT

In this paper, we study the dispersion, ignition and flame characteristics of blended jets of hydrogen and methane (as a proxy for natural gas) at near-atmospheric pressure for a fixed volumetric flow rate, which mimics the scenario of a small-scale unintended leak. A reduction in flame height is observed with increasing hydrogen concentration. A laser is tightly focused to generate a spark with sufficient energy to ignite the fuel. The light-up boundary, defined as the delineating location at which a spark ignites into a jet flame or extinguishes, is determined as a contour. The light-up boundary increases in both width and length as the hydrogen content increases up to 75% hydrogen, at which point the axial ignition boundary decreases slightly for pure hydrogen relative to 75% hydrogen. Ignition probability, a key parameter regarding safety, is computed at various axial locations and is also shown to be higher near the nozzle as well as non-zero at further downstream locations as the hydrogen content in the blend increases. Planar laser Raman scattering is used in separate experiments to determine the concentration of both fuel species. Mean fuel concentrations well below the lower flammability limit are both within the light-up boundary and have non-zero ignition probabilities.

1.0 INTRODUCTION

Hydrogen, a carbon-free fuel, blended with natural gas is of interest due to its potential to reduce the carbon intensity and provide a pathway to a sustainable energy future. The storage and transportation of large-volumes of hydrogen is a key challenge and pipelines can be part of a solution with natural-gas pipelines serving as a model system. The United States has a well-established natural gas infrastructure spanning about 3 million miles. Blending hydrogen with natural gas in the existing infrastructure can provide immediate large-scale storage and transportation while lowering the carbon intensity of the natural gas, as well as proving concepts as a transitional technology to a pure hydrogen pipeline network. These gas lines are susceptible to leakage from broken welds, flanges, cracks etc., and the possible ignition of leaks of blends is a safety concern. The ignition probability of blends may increase with an increase in concentration of hydrogen in the mixture, since hydrogen has very low ignition energy. A good understanding of the ignition and flame properties of blends is needed to provide a basis for safety, codes, and standards, which are otherwise applicable only for compressed natural gas.

Figure 1 shows the possible outcomes when a flammable jet encounters a spark. The top row demonstrates a non-ignition event wherein a spark forms, but the fuel remains unignited. This will occur in a region that is too rich or too lean. The middle row shows a failed light-up event in which there is an ignition immediately following the spark, but the kernel extinguishes. This occurs when there is a pocket of fuel with the right stoichiometry to burn, but scalar dissipation is too high to support flame propagation from the kernel. The bottom row shows a situation in which the ignition is followed by kernel expansion which leads to a self-sustained diffusion jet flame. The possibility of having any one of the three events varies within the flow domain, and the ignition probability is used to map out locations where light-up occurs.

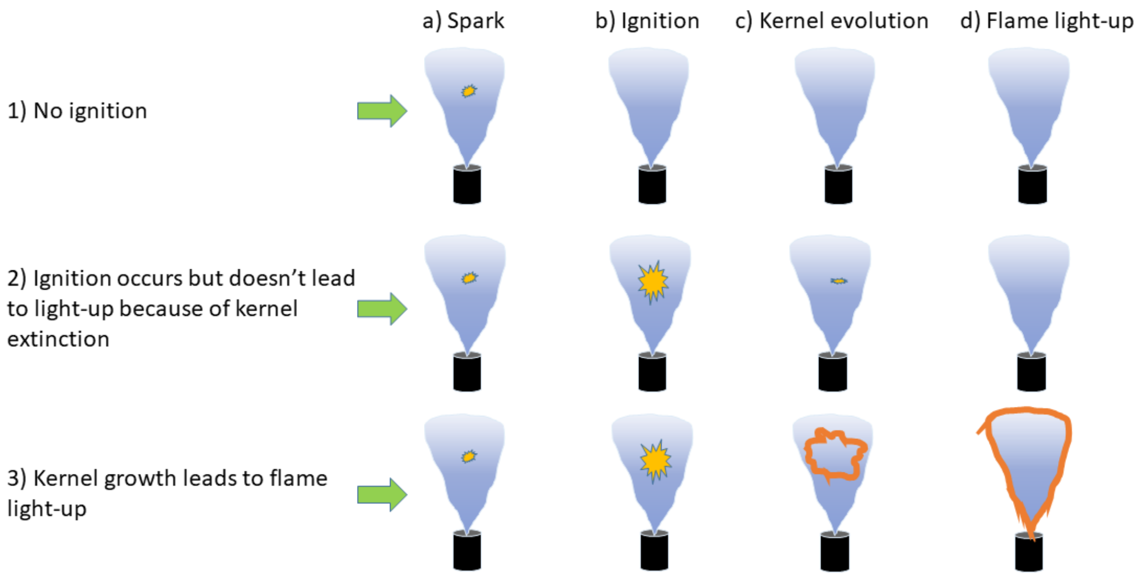

Figure 1. Possible events when a flammable jet encounters a spark: top row illustrates no ignition, middle row ignition with no light-up, bottom row light-up.

Flame length is an important parameter in understanding the hazard and consequences of an unintended gas release event followed by ignition. Since gaseous fuels are often stored at high pressure, the gas jet during a leak event can be under-expanded and flames from even small leaks can reach several meters depending on the tank pressure. Hawthorne et. al. [1] conducted pioneering work on this topic and reported that the flame length is proportional to nozzle diameter only, and the fuel flow rate has no influence on the flame length as long as it is great enough to produce a fully developed turbulent flame, based on their experiments using a wide variety of fuels. On the other hand, Kalghatgi [2] proposed flame length correlations after finding that flame lift-off height varies linearly with the jet exit velocity and is independent of the burner diameter for a given gas. Another important factor that affects the flame length is the nozzle geometry. A study by Henriksen et al. [3] using a complex geometry nozzle suggested that nozzle geometry can increase the flame length by 62% compared to a typical nozzle configuration with equal mass flow rate. Most of the research reported on this topic is conducted using hydrogen [4-6], methane and propane to some extent, with limited data on hydrogen-methane blends [7].

In this work, we characterize the flame length for five mixtures ranging from pure methane to pure hydrogen. We characterize the light-up boundary as well as the ignition probability for these blends. The unignited concentration fields are also mapped out and compared to the light-up boundary and ignition probabilities. This dataset begins to provide much needed information in a previously understudied area.

2.0 EXPERIMENTAL SETUP

The experiments were conducted at Sandia National Laboratories using the setup elucidated in

Figure 2. An electronic mass flow controller is used to meter the gas flow rate into a settling chamber, which is subsequently released into the ambient through a circular orifice of 1 mm diameter. A 3-axis linear translation stage is used to move the release point, enabling measurements at different heights or to change the relative position of the spark to the release point. All the gas valves, fuel flow meters and pressure regulators are controlled using a LabView program.

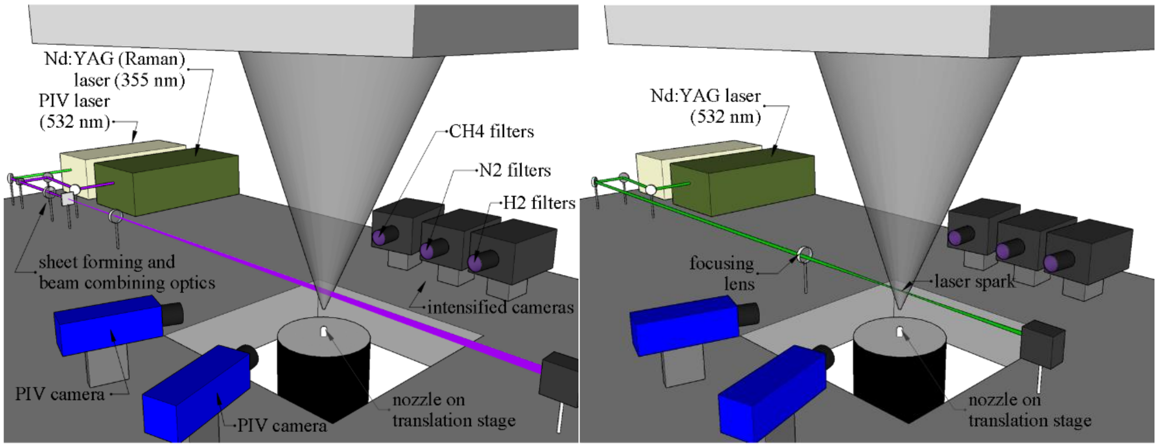

Figure 2. Experimental setup during planar laser Raman experiments (left) and igntion experiments (right).

To measure the light-up boundary and ignition probabilities at various radial and axial locations without disturbing the flow, a laser-based ignition system shown on the right of

Figure 2 is used. The 532 nm output beam from a pulsed Nd:YAG laser is focused into a tiny spot using a spherical lens leading to optical breakdown followed by plasma formation. The energy of the laser beam (typically around 600 mJ/pulse) is well above the minimum ignition energy for both hydrogen and methane.

In addition to understanding the light-up boundary, quantitative measurements of H_2 and CH_4 concentrations were performed using planar laser Raman scattering with the setup shown on the left of

Figure 2. The third harmonic of a pulsed Nd:YAG laser (Continuum PL-9010) with a pulse width of 7 ns and approximately 450 mJ/pulse provided sufficient energy to excite various species of interest present in the flow. Spherical and cylindrical lenses were used to shape the laser beam into a sheet. Raman signals from the molecules of interest (viz. hydrogen, methane and nitrogen) were captured using intensified CCD cameras. These cameras were outfitted with corresponding filters (420 ± 10 nm for H_2, 387 ±10 nm for N_2, 400 ± 10 nm for CH_4) and 355nm notch filters (OD 6) to restrict other wavelengths and to remove the Rayleigh and background scattered light.

3.0 TEST CONDITIONS

Experiments were conducted with a constant fuel flow rate of 0.75 slm through a 1 mm diameter orifice for the gas compositions detailed in Table 1. The flow exits the orifice at 16 m/s which corresponds to Reynolds numbers of 147 and 947 for hydrogen and methane respectively. The flow velocities were kept low since preliminary experiments showed poor flame stability of methane at high flow velocities downstream of the orifice.

Table 1. Test conditions.

Gas Composition	Flow rate, slm	Orifice Diameter
100% H_2	0.75	1 mm
75% H_2-25% CH_4	0.75	1 mm
50% H_2-50% CH_4	0.75	1 mm
25% H_2-75% CH_4	0.75	1 mm
100 % CH_4	0.75	1 mm

4.0 RESULTS

4.1 FLAME HEIGHT MEASUREMENTS

Jet flame height is an important parameter for safety, as it is related to the convective and radiative output and is sometimes used to quantify the magnitude of discharge [8]. Here, the flame height is defined as the distance along the vertical axis (z-axis) from the exit of the nozzle to the tip of the flame. This includes any standoff distance from the nozzle; for low flow rates the flames studied here were typically anchored on the nozzle. To measure this distance, we recorded the flame images for a duration of 200 seconds using a CCD camera. The flame edges were derived by using a Canny edge detection algorithm, and the flame length was averaged for the 2000 images.

Figure 3 shows the mean flame heights for the pure fuels and the different hydrogen-methane blends. With increase in hydrogen content of the fuel, there is a monotonic, nearly linear reduction in the flame height. This is because of the increase in flame speed due to hydrogen addition, which ranges from around 40 cm/s for pure methane to 210 cm/s for pure hydrogen [9]. The increased flame speed causes an increase in consumption of the fuel with hydrogen addition (i.e., the products are consumed nearer to the nozzle) and the flame is confined to shorter region.

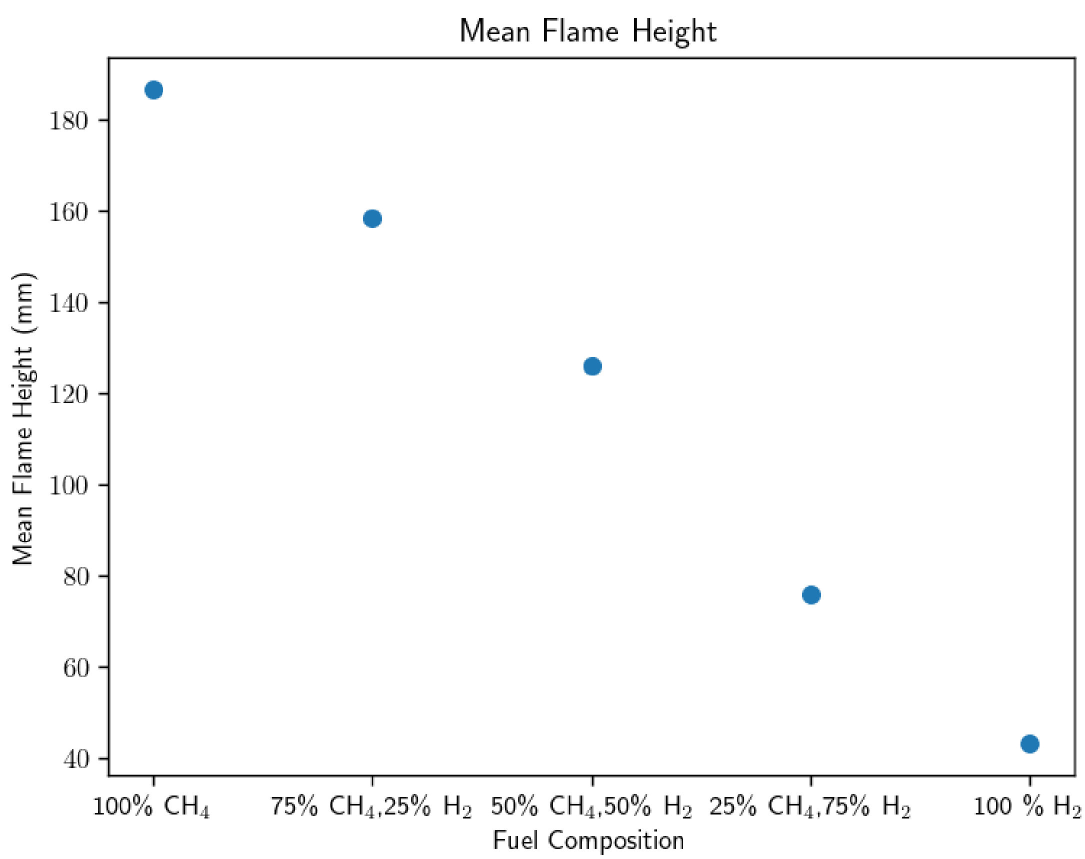

Figure 3. Mean flame height for various H_2-CH_4 blends with a constant fuel flow rate of 0.75 slm exiting through a 1 mm diameter orifice.

4.2 LIGHT-UP BOUNDARY

As shown in the second two rows of Figure 1, after the successful initiation of a flame kernel, there exists two possibilities, the first one being the expansion of ignition kernel and the upstream propagation of the flame leading to light-up (bottom row), or kernel extinction (middle row). The light-up boundary is defined as the spatial location that separates these two regions [10]. In the present study, the light-up boundary is determined at a particular height by moving the experimental setup such that the ignition kernel location moves radially outwards. After every translation, sufficient delay is provided for the jet to stabilize before the next run. Figure 4 shows the light-up boundary for methane, the different blends, and hydrogen, obtained by averaging three independent measurements of the boundary profiles and for each boundary profile measurement, an average of five tests at each height.

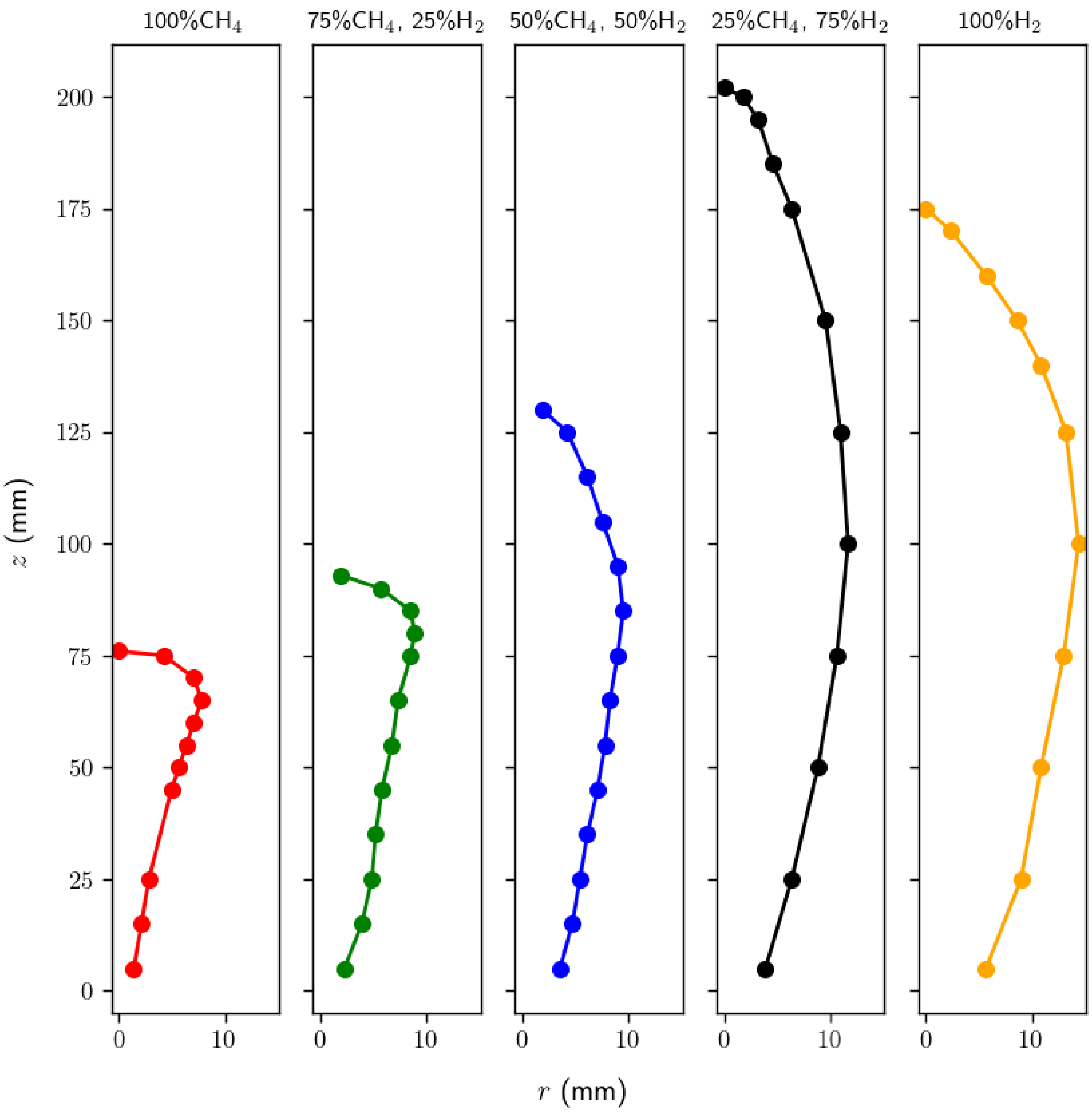

Figure 4. Flame light-up boundary of methane, blended, and hydrogen jets with constant fuel flow rate of 0.75 slm exiting through a 1 mm diameter orifice.

Figure 4 shows that both the radial and axial dimensions of the light-up boundary increase with the percentage of hydrogen in the blend, with 100 % H_2 being an exception regarding the maximum jet height. As shown later in Section 4.4, higher mole fractions of methane remain further downstream than hydrogen, potentially due to the diffusional differences between methane and hydrogen (hydrogen diffuses more rapidly radially, lowering the concentration axially). The methane in the 25% CH_4, 75% H_2 blend is likely penetrating further downstream, with a small content of hydrogen which increases the flame speed for lean mixtures, extending the axial extent of ignition probability.

The ignition boundary trend is nearly opposite of the flame length trend, also due to the flame speed. In lean downstream or far radial regions for mixtures with a low flame speed (i.e., with more methane), the flow momentum will blow the kernel downstream, leading to extinction. If the flame speed is higher (i.e., blends with more hydrogen), the flame propagation speed is high enough that the reaction can propagate against the flow momentum and ignite into a jet flame.

4.3 IGNITION PROBABILITY

Ignition probability is an important metric that determines the possibility of forming an ignition kernel when a mixture encounters a spark. In this case, ignition is considered to have occurred whether the kernel extinguishes or propagates into a jet flame (either row ii or iii from Figure 1). For measuring ignition probability, the same translation protocols used for light-up boundary determination are used. The laser is fired continuously at 10 Hz and a photodiode is used to register all the pulses. A photomultiplier tube (PMT) is focused on the ignition zone, which sees the laser breakdown and a possible ignition event afterwards. By comparing the signals from the PMT and photodiode, based on a common reference point, the pulse corresponding to ignition can be identified. Ignition probability is calculated as the inverse of the number of pulses elapsed before a successful ignition for each trial, and several trials are averaged to obtain its final value. The ignition location coordinates (r,z) are (0, 75 mm).

The ignition probabilities along the centerline for all the conditions mentioned in Table 1 are shown in Figure 5. In the region close to the orifice exit, pure hydrogen and the blends with a hydrogen mole fraction ≥ 50% have an ignition probability of 100% while methane has a lower probability (around 40 %) because of insufficient mixing and the much lower upper flammability limit for methane (15%) than hydrogen (75%). Once sufficient mixing has occurred, the ignition probability of methane increases up to 94 % (at $z = 60$ mm) before dropping back to zero (at $z = 140$ mm).

As evident from Figure 5, the ignition probability increases with additional hydrogen content. The blends with 50% or more hydrogen content have an ignition probability of 100% along the centerline up to an axial location which varies between $5 \leq z \leq 75$ mm. For pure hydrogen, a centerline ignition probability of 100% is observed up to z = 150 mm. It should be noted that the farthest point along the axis at which a successful ignition is observed doesn't correspond to pure hydrogen but rather 25% methane, 75% hydrogen. This was a similar trend regarding the axial flame light-up boundary shown in Figure 4 where this mixture had a light-up boundary that extended further downstream than for pure hydrogen. Once again, a potential explanation is that the less diffusive and heavier methane penetrates further downstream than the hydrogen. It should be noted that the ignition probability is low (<20%) but not zero more than 150 mm downstream. The ignition probability is an important parameter in risk calculations, since the hazard associate with a flame are much different than the hazards associated with unignited dispersion.

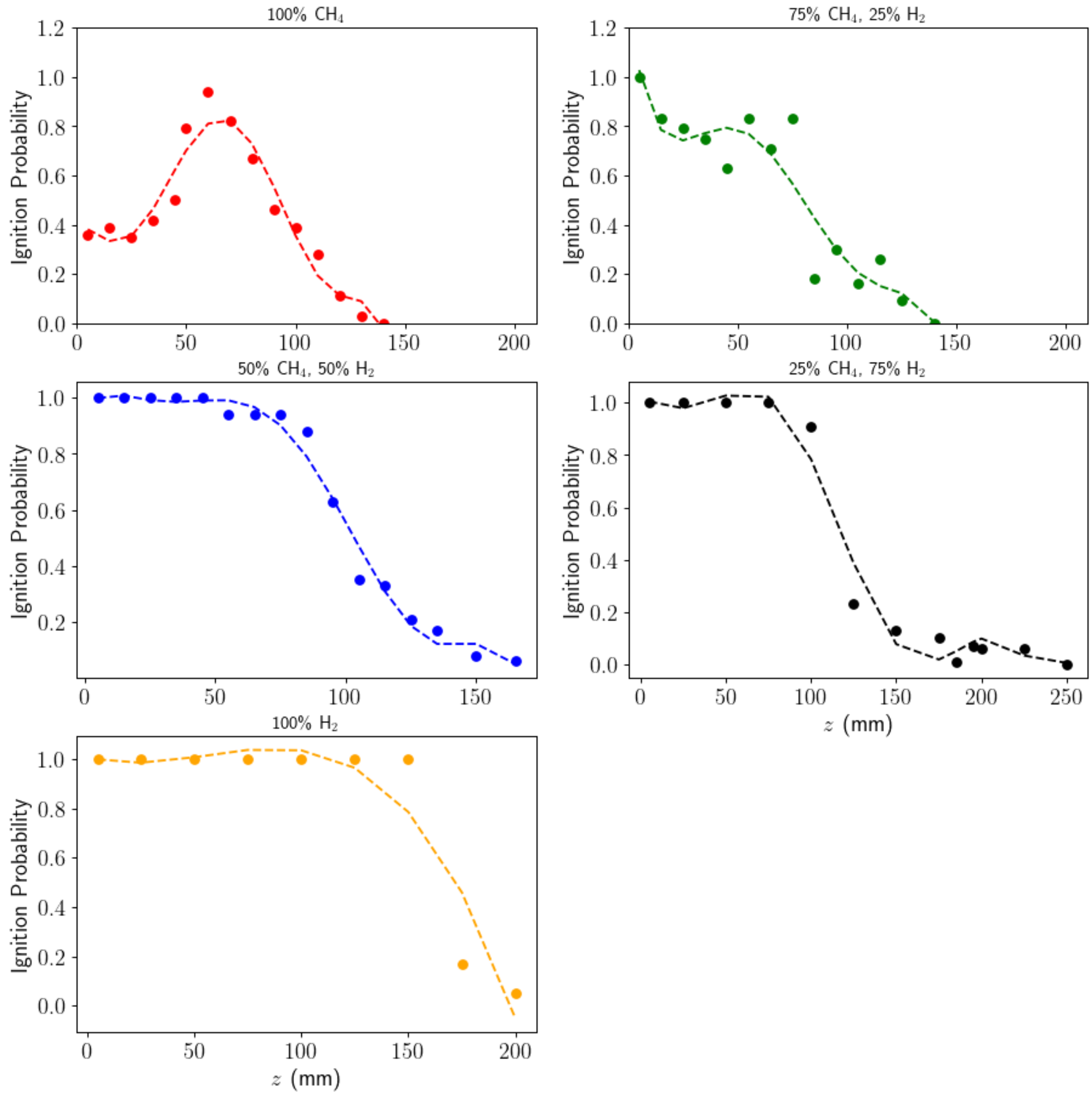

Figure 5. Axial ignition probability of methane, blended, and hydrogen jets.

4.4 MOLE FRACTION MEASUREMENTS

Quantitative measurements utilizing a planar laser Raman scattering technique was conducted to acquire two-dimensional mole fraction fields of hydrogen and methane for the various blends mentioned in Table 1. In the region immediately downstream of the orifice, laser reflections are prominent and hence the data is acquired from a location starting $z = 16$ mm. In order to achieve a good signal-to-noise ratio, the laser beam is expanded into a sheet of approximately 30 mm, of which 25 mm has sufficient power to observe spontaneous Raman scattering. Multiple regions are imaged by capturing 400 images at 10 Hz, extending up to a height of 210 mm.

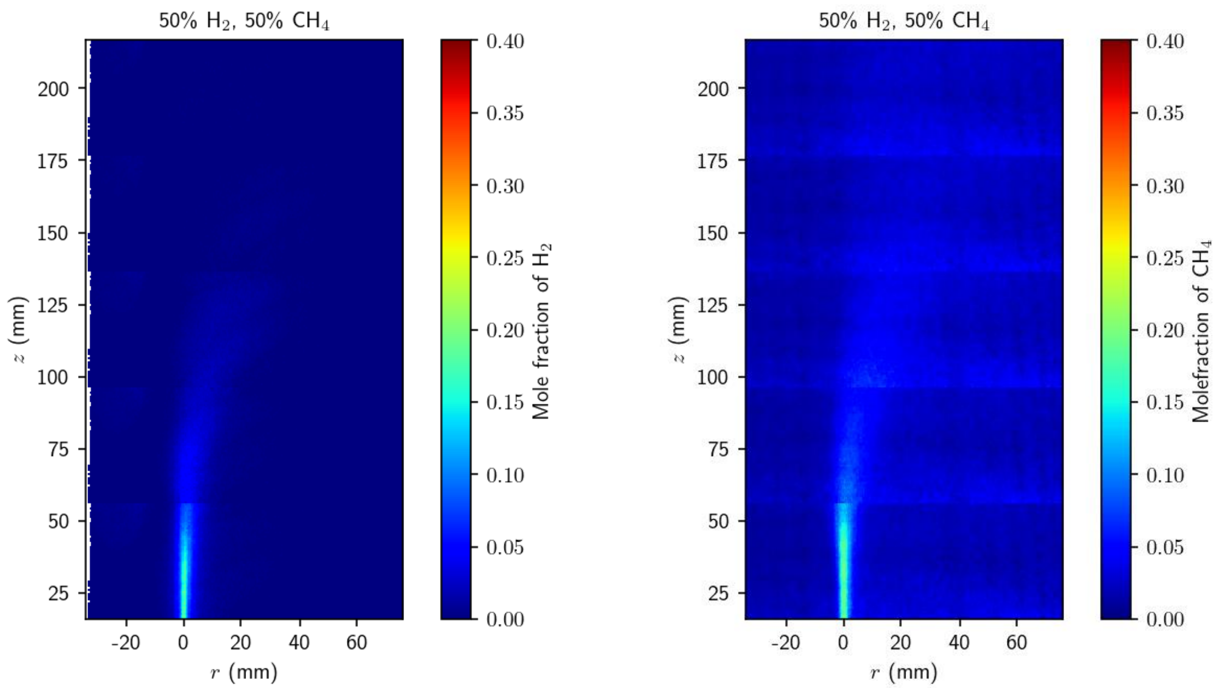

Figure 6. Hydrogen (left) and methane (right) mole-fraction fields for the 50% H_2, 50% CH_4 mixture.

The median mole fractions of the 400 images at each axial location are stitched together to obtain the full mole fraction field; the hydrogen and methane mole fraction fields for the 50% H_2-50% CH_4 blend are shown in Figure 6. The air region in the stitched image for methane still has some signal, particularly along the bottom edge of each imaging plane. This appears as the 5 horizontal streaks across the image. This is an artifact of the data processing, which can be improved by filtering as described by Li et al. [13] in future work. Both the hydrogen and methane mole fraction fields show the jets tilting towards the right-hand side of the image more than 50 mm downstream. This is likely caused by the active ventilation system above the releases or other air currents in the room during the release experiments. Jet tilting appears to be a function of release velocity and is not taken into account in the present study; the reported centerline or axial characteristics are vertically above the release point.

Figure 7 shows the evolution of centerline mole fraction profiles of hydrogen and methane for all of the experimental conditions, as listed in Table 1. It should be noted that the simultaneous measurements of hydrogen and methane mole fraction fields are limited only to the blends, while the 100% hydrogen and 100% methane cases are measured separately (i.e., there is no signal for methane in the 100% hydrogen case and vice-versa). The concentration profiles exhibit a hyperbolically decaying trend, which is consistent with the results reported in the literature [11-12] along with a separate work on high pressure blends submitted to this conference by the authors. The concentration gradient is maximum in the near vicinity of the orifice. The flat to increasing mole fractions between 16 and 30 mm downstream are artifacts from noise during the data processing. We expect the mole fractions to monotonically decrease with improved data processing. Along the axis, the species mole fractions decrease rapidly due to the entrainment of ambient air and asymptotically approach zero in the far field region. Notably, the mean concentrations are well below the lower flammability limit (4% for hydrogen and 5% for methane) where the ignition probabilities are non-zero. For example, inspection of Figure 5 shows that the 50% H_2, 50% CH_4 blend has non-zero

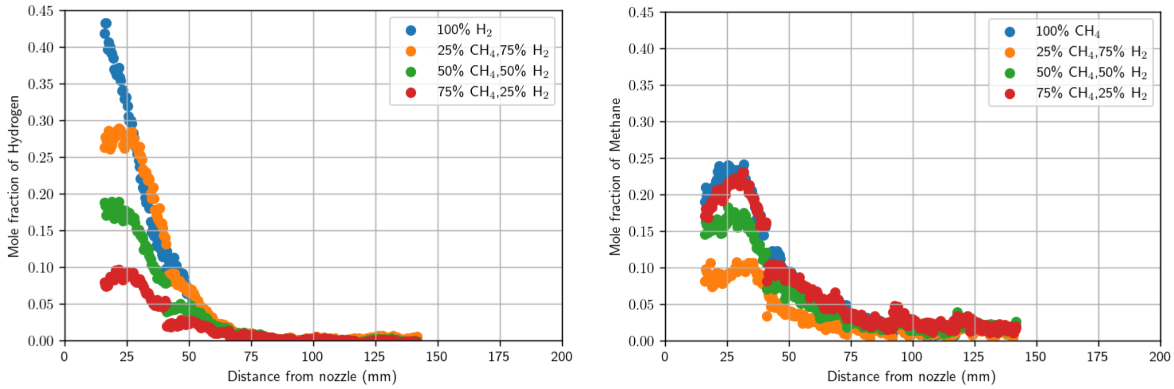

Figure 7. Centerline mole-fractions of hydrogen and methane for different blends at 0.75 slm

ignition probability all the way out to 170 mm. This indicates that the turbulent fluctuations in the flows are significant, bringing pockets of flammable mixtures well downstream of where the average concentration has dropped below the lower flammability limit. The light-up boundaries shown in Figure 4 also extend axially to regions of the flow well below the mean lower flammability limit, contrary to observations by Schefer et al. [14] who found the axial extent of the light-up boundary to be in a region well above the lower flammability limit for both methane and hydrogen. This may be due to the lower velocity of our jets (16 m/s) relative to those in Schefer et al. [14] (134 m/s for hydrogen and 29 m/s for methane).

5.0 SUMMARY AND CONCLUSIONS

In this paper, we present some preliminary results on the dispersion, ignition, and combustion characteristics of hydrogen-methane blends at a constant 0.75 lpm from a 1 mm diameter orifice into the ambient. Chemiluminescence images were captured using a CCD camera, from which the flame height was computed. The mean flame height is inversely proportional to the concentration of hydrogen in the mixture. A spatial contour, termed as light-up boundary, that separates the ignition kernel expansion zone and the kernel annihilation zone was measured by attempting to ignite the flow using a non-intrusive laser based ignition system and measuring the subsequent chemiluminescence with a photomultiplier tube. Both the radial and axial dimensions of the light-up boundary increase with the percentage of hydrogen in the blend, up to 75% H_2, then the axial extent of the light-up boundary slightly decreased with 100% H_2. Another important metric called the ignition probability that quantifies the risk of ignition was measured along the centerline. For 100% H_2 and those blends with 50 % or more hydrogen, the ignition probability is 100% near the nozzle while methane showed a lower value (approximately 40%). Further downstream, the ignition probability of methane increased, peaking at 94%, 60 mm from the nozzle. The ignition probability increases with an increase in hydrogen concentration. A quantitative measurement of the species concentrations using planar laser Raman scattering revealed that the concentration profiles exhibit a hyperbolically decaying trend and asymptotically approach zero, as expected.

In future work, we will reprocess the Raman data with improved filtering to decrease the noise. We also plan to measure the velocities of these flows using particle imaging velocimetry. We plan to develop an ignition model for blends (as well as pure methane and hydrogen) based on mean and fluctuating concentration and velocity information. This model can be used for risk assessments and the development of safety codes and standards for blends of natural gas and hydrogen and increase the accuracy and probabilistic ignition modeling for hydrogen.

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