**Simultaneous formaldehyde PLIF and high-speed schlieren imaging for
ignition visualization in high-pressure spray flames**

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Abstract:

We applied simultaneous schlieren and formaldehyde (CH_2O) planar laser-induced fluorescence (PLIF) imaging to investigate the low- and high-temperature auto-ignition events in a high-pressure (60 bar) spray of n-dodecane. High-speed (150 kHz) schlieren imaging allowed visualization of the temporal progression of the fuel vapor penetration as well as the low- and high-temperature ignition events, while formaldehyde fluorescence was induced by a pulsed (7 ns), 355-nm planar laser sheet at a select time during the same injection. Fluorescence from polycyclic aromatic hydrocarbons (PAH) was also observed and was distinguished from formaldehyde PLIF both temporally and spatially. A characteristic feature previously recorded in schlieren images of similar flames, in which refractive index gradients significantly diminish, has been confirmed to be coincident with large formaldehyde fluorescence signal during low-temperature ignition. Low-temperature reactions initiate near the radial periphery of the spray on the injector side of the spray head. Formaldehyde persists on the injector side of the lift-off length and forms rapidly near the injector following the end of injection. The consumption of formaldehyde coincides with the position and timing of high-temperature ignition and low-density zones that are clearly evident in the schlieren imaging. After the end of injection, the formaldehyde that formed on the injector side of the lift-off length is consumed as a high-temperature ignition front propagates back toward the injector tip.

Keywords: spray combustion; diesel combustion; high-speed imaging; formaldehyde PLIF; schlieren imaging

1. Introduction

Ignition characteristics and flame stabilization represent two of the critical factors considered in the design and operation of compression ignition engines as they determine operating limits and influence pollutant formation. As such, it is imperative that combustion models accurately reproduce experimental ignition delay time and lift-off length measurements. Modeling efforts from various research institutions, however, show inconsistency between experimentally determined and modeled ignition delay times and lift-off lengths [1-6]. To improve the current understanding of ignition and stabilization processes in high-pressure spray flames and enable the development of more accurate models, information is needed about the temporal evolution of the spray as it progresses through low- and high-temperature ignition stages and subsequently stabilizes as a lifted diffusion flame.

In compression ignition engines, low-temperature chemical reactions yielding radicals and reactive intermediates precede the high-temperature ignition event and may play an important role in subsequent stabilization of the quasi-steady lifted flame [7, 8]. While some of the aforementioned species can be visualized using planar laser-induced fluorescence (PLIF), limitations associated with lasers and/or intensified cameras have constrained most such measurements to a single-shot per injection event. Using an array of lasers to generate a train of pulses and a framing camera, researchers have acquired up to eight sequential time-resolved images of fuel-, formaldehyde-, and OH-PLIF spaced 10 μ s apart during a single engine cycle [9, 10]. Recent developments in high-speed burst lasers and high-speed intensified cameras have also permitted PLIF imaging measurements at rates as high as 12 kHz with a single laser and camera setup [11]. Such instrumentation can be cost-prohibitive, however, and has not yet been

widely used throughout the engines and spray combustion communities. Moreover, even at 12 kHz critical events during the ignition process can be missed. In the present work, we apply simultaneous high-speed schlieren imaging at 150 kHz and single-shot formaldehyde PLIF at various timings as a simpler alternative to high-speed PLIF for visualizing the temporal evolution of a reacting spray.

Several research groups have used high-speed schlieren imaging to visualize low-temperature ignition processes in diesel sprays [12-15]. In those experiments, the low-temperature ignition event was observed as a local “softening” of refractive index gradients near the spray head. This phenomenon was attributed to the consumption of fuel vapor resulting in the production of intermediate species and heat release that increased the local temperature to a value closer to that of the ambient gases. Consequently, the local refractive index value approached that of the surrounding gases rendering the schlieren effect less pronounced. As formaldehyde (CH_2O) has been observed by PLIF imaging during the low-temperature ignition event, it was supposed that the softening of refractive index gradients in the schlieren imaging would be coincident with regions of formaldehyde and that the formaldehyde would be consumed following high-temperature ignition. The present study investigates this hypothesis by performing high-speed schlieren with simultaneous single-shot formaldehyde PLIF from the same perspective.

2. Experimental Setup

2.1 Constant Volume Pre-burn Combustion Vessel

Sprays of n-dodecane ($C_{12}H_{26}$) were injected into a pre-burn combustion vessel that simulates the thermodynamic conditions of modern compression-ignition engines [15, 16]. A common-rail single-hole diesel fuel injector (# 210370, 90- μm orifice) belonging to the family of Spray A injectors of the Engine Combustion Network (ECN) [15] injected fuel at 1500 bar for approximately 1.5 ms. The injection duration and ambient condition for reacting sprays in this work corresponds to the ECN Spray A target (i.e., 900 K, 22.8 kg/m³, and 15 vol.% O₂). For the non-reacting cases investigated, the target ambient oxygen concentration was zero.

2.2 Optical Diagnostics

A large beam-splitter (Edmund Optics #43-362) enabled simultaneous PLIF and schlieren imaging of the combustion event from the same perspective as shown in Fig. 1. Images captured from the same perspective allow for a direct comparison of flame structures observed from both diagnostics, which are discussed in detail below.

2.2.1 Formaldehyde PLIF Imaging

Radiation at 355 nm (100 mJ/pulse), produced by a 10-Hz (7-ns pulse) Quanta-Ray Nd:YAG laser and formed into an 80-mm $\times$ 0.3-mm sheet, excited the formaldehyde fluorescence. The laser sheet intersected the fuel jet from the bottom of the vessel and passed through the jet centerline as shown in Fig. 1. A flat field correction for the laser intensity profile along the spray axis, which would result in a more even representation of the fluorescence signal, was not made; however, we do not believe this affects the qualitative comparisons of the PLIF

signal at different timings ASOI. An intensified CCD camera (Princeton Instruments, PI-MAX3) with a 250-ns gate time imaged the PLIF signal. The camera was equipped with a 50-mm f/1.2 Nikkor glass lens along with a 450-nm short-pass filter and a 385-nm long-pass filter to block flame luminosity, elastically scattered light, and reflections from the 355-nm excitation, while transmitting over the strongest resulting fluorescence bands of formaldehyde [17].

Formaldehyde PLIF as described above can potentially suffer from interference due to polycyclic aromatic hydrocarbon (PAH) fluorescence as well as laser-induced incandescence (LII) from soot; however, optical engine and spray vessel experiments have leveraged formaldehyde's characteristic spectral features to demonstrate that formaldehyde fluorescence can be distinguished from PAH and soot interference both temporally and spatially [8, 18]. O'Connor and Musculus [19] also concluded that fluorescence from formaldehyde and PAH in a diesel spray flame could be distinguished by considering their relative intensities, with the formaldehyde fluorescence yield notably smaller than that attributed to PAH. This simplified approach is valid provided that the laser fluence is below the saturation threshold for PAH-LIF [20].

With the above considerations, we note that the operating conditions of the present work promote both temporal and spatial separation of the formaldehyde LIF from PAH-LIF and soot-LII. Finally, we note that the formyl radical (HCO) may also be an indicator of cool-flame ignition; however, it is not anticipated to yield fluorescence signal at the excitation wavelength used in this study. Moreover, calculations based on detailed chemical models show that formaldehyde is the predominant species apparent in these low-temperature reactions.

To portray the expectations for formaldehyde formation during low- and high-temperature ignition, we used an open reactor model that mimics the mixing rates of a fuel jet into an oxidizer [21]. This model has been applied to represent diesel spray conditions (e.g. Spray A conditions), including mixing rates without a diffusion flame and prior to high-temperature ignition [22]. The calculated mole fractions of the parent fuel, formaldehyde, and HCO are presented in Fig. 2 as a function of temperature. The fuel mole fraction has been reduced by a factor of 100, while the HCO mole fraction has been multiplied by 1000. The bounded double-headed arrows indicate the time elapsed between select temperatures (800, 1100, and 1500 K) attained within the reactor. Nearly 500 μ s elapses during the low-temperature reactions as the temperature rises from 800 K to 1100 K. The rise in temperature from 1100 K to 1500 K occurs in 55 μ s, almost a factor of 10 faster. The results from this model also indicate that the mole fraction of formaldehyde is more than three orders of magnitude larger than that of HCO during the low-temperature ignition event. Consequently, the LIF signal from the cool flame will be referred to as formaldehyde LIF.

2.2.2 High-speed schlieren imaging

High-speed imaging of the vapor penetration, cool-flame ignition, and high-temperature ignition were made using a Z-type schlieren setup [23] in an arrangement similar to Ref. [24]. A high-speed CMOS camera (Photron SA-X2) imaged the spray at 150,000 frames per second with a 2- μ s exposure time. An 85-mm f/1.8 Nikkor glass lens resulted in a projected pixel size of 0.213 mm/pixel and a 628-nm (32 nm FWHM) band-pass filter was mounted on the lens to reject flame luminosity. A custom LED, emitting 3.5 W of 635 nm (15 nm FWHM) radiation in 1- μ s pulses, was synchronized with the high-speed camera to provide illumination for the schlieren

setup. Using a short LED pulse further enhanced temporal resolution. A 50-mm condensing lens collected the LED emission and directed it through a 3-mm aperture prior to collimation by a 115-mm diameter, f/8 parabolic mirror. After passing through the combustion vessel, the collimated beam was re-focused using a second parabolic mirror and a series of flat folding mirrors. The light was imaged by the high-speed camera approximately 100 mm after the focal point, where a schlieren stop aperture of 12-mm diameter was placed to yield the desired contrast to density gradients. The 3-mm aperture at the light source and the 12-mm schlieren stop aperture were selected by trial and error to reduce the intensity of unwanted background schlieren caused by temperature gradients at the windows, while maximizing the contrast of softening gradients during the low-temperature ignition event similar to Pastor et al. [14]. To further reduce the appearance of background schlieren structures, the schlieren images were processed by normalizing the images by a frame just prior to the beginning of injection.

3. Results and Discussion

Previous research using high-speed schlieren imaging has observed a sudden softening of the characteristic refractive index gradients just prior to the high-temperature ignition event giving the appearance of “transparent” schlieren from the fuel jet [12, 13]. Similarly, high-speed imaging of the broadband flame emission has demonstrated the appearance of low-level chemiluminescence at the same timing prior to high-temperature ignition [12, 18]. It was hypothesized that these phenomena were associated with the low-temperature auto-ignition chemistry forming the necessary radicals to initiate high-temperature ignition as well as intermediates such as formaldehyde. The results presented below demonstrate that the low-

temperature heat release or “cool-flame” event is indeed responsible for the softening of refractive index gradients prior to the high-temperature ignition event.

3.1 Schlieren imaging of reacting and non-reacting fuel sprays

To confirm that the softening of the schlieren effect is associated with low-temperature reactions, schlieren imaging was performed under both non-reacting and reacting conditions. Images from both cases at select timings after the start of injection (ASOI) are presented in Fig. 3. The non-reacting spray is shown in the panels on the left side of the figure, while the reacting spray results are shown on the right. The timing ASOI associated with each image is shown in the upper left corner of each image. High-speed movies and LIF images acquired in the present study have been made available on the ECN website [25].

In the top two frames at 114 μs and 260 μs ASOI, the appearance of the non-reacting and reacting sprays is quite similar and the dark regions induced in the image by the liquid and vaporizing fuel are easily distinguished from the ambient gases and background structures. With regard to fuel penetration, the axial penetration and the width of the radial boundaries at both of these early timings are within a few millimeters of each other for the non-reacting and reacting cases. At this time, it is important to point out that the liquid boundary ends at an axial distance of approximately 10 mm for both cases under these conditions; thus, at axial positions beyond 10 mm the schlieren effect is induced by refractive index gradients between fuel vapor and the ambient gases. In the third panel from the top, at 280 μs ASOI, the schlieren image for the non-reacting case has not significantly changed relative to the previous time step; however, the reacting case begins to show a “softening” effect near the head of the spray. More specifically, at the previous timing (260 μs ASOI), the lower portion of the “reacting” spray head is

characterized by a dark semi-circular region (shown outlined in white for clarity). In the following frame (280 μs ASOI), this same structure is visible (also outlined in white); however, the region appears lighter. This is a consequence of reduced gradients in the local refractive index, which result from a rise in the local temperature to values closer to the temperature of the ambient gases. This temperature rise can be attributed to heat release from the low-temperature auto-ignition reactions [24]. A similar effect is not observed in the non-reacting case presumably because molecular oxygen is not present in the ambient and low-temperature oxidation reactions cannot occur.

At later timings (314 μs and 347 μs) the non-reacting and reacting sprays look quite different. Whereas the non-reacting spray maintains its radial width and continues to penetrate away from the injector orifice, the head of the reacting spray thins and then becomes transparent. The images at these timing illustrate that the fuel vapor is being consumed from the outer radius toward the central axis. At 347 μs in the reacting case, the low-temperature reactions first observed at the periphery of the spray head at 280 μs have now consumed a significant portion of the fuel vapor within the spray head resulting in a distinctly brighter region between 20 and 25 mm from the injector orifice (compared to the non-reacting case). At 380 μs , high-temperature ignition has ensued in the reacting case, as confirmed by the associated pressure trace and high-speed chemiluminescence imaging under identical conditions, and the axial penetration boundary of the reactive species defined by the flame is consistent once again with the non-reacting vapor penetration. Moreover, faint structures observed in the head of the reacting jet at 347 μs retain their shape when they darken after high-temperature ignition at 380 μs . This provides strong evidence that high-temperature ignition is a volumetric event occurring within the region where low-temperature reactions have produced radical and intermediate species. Finally, in the bottom

right panel at 680 μs the quasi-steady lift-off length has been established in the reacting spray and can be identified by the abrupt radial expansion of the schlieren boundary near 17 mm. This radial expansion of the high-temperature products near the lift-off length has been emphasized by overlaying the non-reacting vapor boundary at this same timing (corresponding to the lower left panel) as a white border.

By inspecting the high-speed schlieren imaging for 18 independent injections, the following characteristic features could be discerned for the flame conditions investigated here. Typically, the schlieren first softens near the radial periphery just behind the penetrating spray head between 17 and 23 mm suggesting that low-temperature chemistry may initiate in this region rather than in the core of the spray. Along the centerline 20 mm from the injector orifice, the Rayleigh scattering measurements of Pickett et al. [26] showed that the equivalence ratio (ϕ) and temperature were approximately four and 720 K, respectively. In contradistinction, at the same axial location in the radial periphery where ϕ is near unity, the local temperature was greater than 850 K. The model results presented in Fig. 2 indicate that formaldehyde formation would not be significant below approximately 800 K. Thus, on average, it appears less likely that low-temperature chemistry would initiate in the core of the spray.

Within 50-100 μs after cool-flame initiation at the radial periphery the entire spray head becomes transparent, suggesting homogeneity in temperature along a line of sight within this region (and with the ambient) and indicating that the parent fuel has decomposed. From these observations it could be hypothesized that reactions initiating in the radial periphery provide the necessary reactive species and heat release to promote the low-temperature chemistry throughout the entire spray head, which subsequently undergoes high-temperature ignition volumetrically.

Musculus et al. [27] simulated the ignition of vapor phase n-heptane at two different temperatures and equivalence ratios representative of two different radial locations in the jet. Cool-flame reactions initiated more than 1.5 ms earlier in the hotter/leaner (802 K, $\phi=0.7$) mixture, representative of the radial periphery, than in the cooler/richer (744 K, $\phi=2$) mixture representative of the core. The time delay between low- and high-temperature ignition, however, was much longer for the hotter/leaner mixture, which provides further evidence that a coupling exists between the periphery and the core in the high-temperature ignition process.

3.2 Simultaneous CH₂O PLIF and schlieren imaging

The images in Fig. 4 represent a time sequence of formaldehyde PLIF and schlieren images resolving the low- and high-temperature ignition events. False color images of the formaldehyde PLIF are presented in the left column and the limits of the color palette are identical for all panels. In this sequence, formaldehyde PLIF is not observed at 140 μ s ASOI and the image at 190 μ s ASOI has been multiplied by a factor of 3 for illustrative purposes. Images from the high-speed schlieren movies are shown in the right column. The white boundary in the schlieren images corresponds to the formaldehyde PLIF as defined by the region in which signal exceeds 10% of the displayed fluorescence intensity palette. For the case at 190 μ s ASOI, this operation was performed after image amplification. Because the formaldehyde PLIF imaging is a single-shot diagnostic, each PLIF image was acquired from an independent spray event. For purposes of comparison, the schlieren images were extracted at the time corresponding to the PLIF image from that same independent injection event. This means that the schlieren image sequence does not represent a time sequence from a single injection as was shown previously in Fig. 3. Though not shown here, PLIF images were acquired at the timings shown in Fig. 4 during

non-reacting spray events. In all cases, only a few counts of signal were detected presumably due to elastic scatter from the liquid droplets. There was no evidence that the n-dodecane fuel fluoresced as a result of the incident 355 nm radiation. Thus, the fluorescence observed in Fig. 4 is entirely attributed to combustion intermediates such as formaldehyde.

In Fig. 3, the first evidence of low-temperature auto-ignition was observed 280 μ s ASOI when the schlieren effect near the radial periphery of the spray head began to “soften”. In Fig. 4, this event is observed in the schlieren image at 240 μ s ASOI where we see two lighter shaded regions on the upper and lower boundaries of the spray between 17 and 20 mm. Nevertheless, we observe fluorescence from formaldehyde 50 μ s earlier (from a different injection event) suggesting that the schlieren diagnostic may not be sufficiently sensitive to detect the earliest low temperature reaction events.

As discussed above, the schlieren imaging indicated that low-temperature chemistry initiates at the radial periphery of the jet rather than in the core. In support of this, at 190 μ s in Fig. 4 we observe distinct upper and lower lobes of formaldehyde fluorescence on the injector side of the penetrating spray head. That is, while we do observe some fluorescence in the core of the spray, the majority of the signal is found near the radial periphery. Once formed, these lobes of formaldehyde may be transported into the core of the spray by rapid convective mixing resulting in fluorescence signal in the spray head. The same rapid mixing may also occasionally form regions within the core of the spray on the injector side of the spray head where the temperature and equivalence ratio are conducive to low-temperature chemistry coincident in time with the reactions observed at the radial periphery. As explained above, small probabilities for these mixtures within the core mean that these small pockets are not expected to dominate cool-

flame initiation and are therefore less likely to be detectable by a line of sight diagnostic such as schlieren, but they may be detectable in the spray center using the planar fluorescence diagnostic.

At 240 μs ASOI, the upper and lower lobes of formaldehyde fluorescence on the injector side of the spray head remain and the location of the peak fluorescence signal clearly corresponds to the region in the schlieren image where refractive index gradients have softened. In the schlieren image at 340 μs , light regions are observed in the upper and lower portions of the spray between 15 mm and 20 mm, while the head of the spray also appears to have softened relative to the core of the spray. At 390 μs ASOI, high-temperature ignition has occurred as indicated by the radially and axially expanding spray head. Nevertheless, the formaldehyde closer to the injector persists here as well as at the later timings shown. The persistence of formaldehyde on the injector side of the lift-off location is consistent with the results of Refs. [7, 8] and suggests that the low-temperature auto-ignition chemistry plays a role in determining the quasi-steady flame location and is an important target for model development. At the latest timing shown in Fig. 4 (490 μs), the fluorescence near 30 mm is likely due to the onset of PAH.

In Fig. 5, we present a time sequence of formaldehyde and PAH PLIF and the corresponding schlieren images at timings later than those presented in Fig. 4. Here, the PLIF and schlieren images have been paired vertically as opposed to horizontally to allow improved visualization of the entire field of view. Consistent with the results O'Connor and Musculus [19], fluorescence from the PAH in the present work is notably more intense than that from formaldehyde. Consequently, a higher limit ($\sim 2\times$) was required for the color palette in Fig. 5 relative to Fig. 4. To permit a qualitative comparison of the formaldehyde PLIF in Figs. 4 and 5, the formaldehyde signal in Fig. 5 (outlined by the dashed white box) was amplified by the

appropriate factor related to the higher color palette limit. Also, the PLIF images at the different timings ASOI in Fig. 5 are presented with identical color palette limits. With the full field of view, the lift-off length can again be visualized by the rapid radial expansion in the top two schlieren images near 17 mm. Focusing on the top PLIF image, we observe spatial separation between formaldehyde emission (centered near 20 mm consistent with earlier timings) and fluorescence from PAH (forming near the spray head between 30 and 40 mm) consistent with Refs. [7, 8]. At the next time step (790 μ s ASOI), PAH fluorescence has intensified while that from formaldehyde has diminished. This can be attributed to the additional time and temperature resulting in a higher concentration of PAH, while the high-temperature combustion near the lift-off length has consumed a portion of the formaldehyde present in this region. In the third set of images from the top (1690 μ s), time has advanced nearly 1 ms from the previous frame and injection has ended. In this image pair, the lift-off length (see schlieren image) and the persistence of formaldehyde on the injector side of the lift-off length (see PLIF image) are clearly distinguished. Moreover, formaldehyde is observed within a few millimeters of the injector tip indicating that low-temperature reactions occur even closer to the injector relative to the lift-off length after injection ends.

The emission observed near 40 mm is likely still attributable to PAH fluorescence; however, at least some of the emission farther than 40 mm from the injector orifice can be attributed to broadband soot particle radiation as confirmed by images acquired without laser excitation and high-speed chemiluminescence imaging under identical conditions. In the bottom two image pairs, high-temperature ignition appears to propagate back toward the injector orifice consuming the residual formaldehyde in the near-nozzle region. The rate at which the high-temperature ignition front progresses back toward the injector in the schlieren imaging exceeds

any physically reasonable flame speed; thus, this phenomenon should not be considered “flashback” by flame propagation. The presence of formaldehyde in this near-injector region prior to this event suggests that this is an ignition event consuming radicals and intermediates that remain from previous low-temperature reactions. Finally, in the bottom PLIF image fluorescence from PAH and broadband radiative emission from soot particles have largely disappeared as the surrounding air has been drawn into the core promoting oxidation.

4. Summary and Conclusions

We investigated the low- and high-temperature ignition events in a high-pressure spray of n-dodecane from the same optical perspective using simultaneous single-shot formaldehyde PLIF and high-speed schlieren imaging. Fluorescence from PAH that can interfere with formaldehyde PLIF was separated both temporally and spatially. This work confirms the hypothesis that the low-temperature ignition event in diesel sprays can be visualized by schlieren imaging due to a softening of refractive index gradients in regions where low-temperature reactions yield reactive species and modest heat release. Schlieren imaging proved to be less capable than PLIF at detecting the earliest appearance of low-temperature ignition as the first “softening” of the schlieren effect was observed between 30 μs and 50 μs after the earliest appearance of formaldehyde PLIF. The high-speed schlieren imaging coupled with single-shot formaldehyde PLIF suggests that low-temperature chemistry initiates at the radial periphery of the jet slightly behind the penetrating jet head. The schlieren images also indicate that high-temperature ignition typically proceeds as a volumetric event (i.e., consuming the low-temperature radicals and intermediates within a relatively large volume nearly simultaneously). Formaldehyde was observed within the region defining the quasi-steady lift-off length and,

following the end of injection, formed on the injector side of the lift-off length nearly reaching the injector tip. Subsequently, high-temperature ignition in this near-nozzle region proceeded as an apparent “flashback” event; however, the spatial progression of the reaction exceeded any physically realizable flame speed.

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