

Probing Species Formed by Pilot Injection During Re-Compression in a Controlled Auto-Ignition Engine by H₂CO LIF and Chemiluminescence Imaging

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ABSTRACT

Pilot injection (PI) during the negative-valve-overlap (NVO) period is one method to improve control of combustion in gasoline controlled auto-ignition engines. This is generally attributed to both chemical and thermal effects. However, there are little experimental data on active species formed by the combusting PI and their effect on main combustion in real engines. Thus, it is the objective of the current study to apply and assess optical in-cylinder diagnostics for these species. Firstly, the occurrence and nature of combustion during the NVO period is investigated by spectrally-resolved multi-species flame luminescence measurements. OH*, CH*, HCO*, CO-continuum chemiluminescence, and soot luminosity are recorded. Secondly, spectrally-, spatially-, and cycle-resolved laser-induced fluorescence measurements of formaldehyde are conducted. It is attempted to find a cycle-resolved measure of the chemical effect of PI. However, not any noticeable correlation between the measured formaldehyde content of the charge and main combustion phasing is observed. This can be explained by the findings that the exhaust gas of the prior cycle and potentially pool fire contribute significantly to the measured formaldehyde content of the charge. Thus, formaldehyde is not a good indicator of the chemical effect of PI on main combustion under these conditions.

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INTRODUCTION

Previous research indicated that pilot injection (PI) during NVO is an effective means of controlling combustion phasing and pressure-rise rate at the low and high load regions, respectively, in Controlled Auto-Ignition and Homogeneous-Charge Compression-Ignition (HCCI) engines [1, 2, 3, 4, 5, 6, 7, 8, 9, 10] and references therein. Generally, main combustion may be affected by this split-injection strategy via both chemical and thermal effects, e.g. [1, 2]. Partial fuel conversion during NVO leads to relatively stable low/intermediate-temperature flame products (H₂O₂, H₂CO, etc.), whereas heat is basically released by high-temperature combustion. However, detailed experimental knowledge on the role of high-and low/intermediate-temperature reactions during NVO, which has been measured in real engines, is still lacking. Thus, it is the objective of this study to probe intermediate species formed by PI by optical diagnostics applied to a gasoline controlled auto-ignition (GCAI) engine.

According to [2], mixture homogenization during NVO appears to be a key factor for the optimization of the competing high-and low-temperature combustion effects, at least for two-stage ignition fuels. This is studied by varying the PI timing in the present work, using a single-stage ignition fuel. The relative amount of PI fuel is also varied, since this is another major control parameter [5, 8, 11].

In the present study, intermediate-temperature combustion during NVO is characterized by probing formaldehyde (H₂CO), which is a typical intermediate species [12, 13]. Formaldehyde was previously measured by laser induced fluorescence (LIF) in Controlled Auto-Ignition and HCCI engines during low-temperature reactions at the onset of main oxidation of two-stage ignition fuels [14, 15, 16]. It was also probed between PI and main combustion by LIF in an HCCI engine, using a two-stage ignition fuel [5]. Based on cycle-averaged formaldehyde images, it was demonstrated that the LIF signal decreased with decreasing amount of fuel injected during NVO,

indicating that low-temperature reactions during NVO also decreased. Thus, cycle-resolved formaldehyde data may also show the influence of combustion during the NVO period in terms of “cool”-flame products on cycle-to-cycle variations of main combustion. Therefore, cycle-resolved formaldehyde-LIF measurements are conducted in the present study. This requires a high sensitivity of the optical detection system, which is described in the Experimental section.

It is a major objective of this work to study operating conditions with increased cycle-to-cycle variations close to the edge of the range of feasible engine operation. In contrast to [5, 14, 15, 16], a neat single-stage ignition fuel (iso-octane, purity > 99 %) is used in the present work. Thus, main combustion and resulting formaldehyde formation start late in the compression stroke, when intermediate temperature is reached [17].

Therefore, the formaldehyde produced by PI (or the previous main combustion) can be measured selectively relatively late in the compression stroke, e.g. at -90°CA (crank angle with regard to top-dead center of main combustion). This is beneficial because the gas mixture is approximately homogeneous at this late measurement time. Thereby, the probed gas mixture is almost representative of the entire gas in the cylinder, in spite of the limited size of the probe volume.

In this work, spectrally resolved formaldehyde-LIF measurements are also conducted to identify any potential interfering light emissions, such as LIF from other species and surface-scattered laser radiation. In particular, formaldehyde-LIF may be affected by LIF from poly-cyclic aromatic hydrocarbons (PAHs) [12, 18]. For instance, particularly high PAH emissions were found by speciated-hydrocarbon exhaust-gas analysis applied to a two-stroke HCCI engine [19]. On the other hand, the PAH content of the exhaust gas appeared to be relatively low for the current GCI engine, at least for single-injection fueling strategies, as described in a companion paper [20]. Thus, the PAH content of the currently probed gas mixtures is unclear. This is a major reason for the present spectrally-resolved LIF measurements.

Furthermore, combustion during NVO is characterized by spatially and spectrally resolved chemiluminescence imaging. This is described in more detail below.

The previous research on reactive species formed during the NVO period was recently reviewed in [21]. Briefly, CO, H₂ and a number of hydrocarbons, such as methane, ethane, ethylene, propene, acetylene, 1,3-butadiene, methanol, acetaldehyde, and formaldehyde were identified. This was conducted by both computations and experimental techniques, such as in-cylinder gas sampling and Fourier transform infrared absorption [1, 2, 3, 21, 22] and references therein. According to [1, 2, 23], hydrogen peroxide (H₂O₂) may be a particularly important species formed by PI, since H₂O₂ represents a temporary storage system for OH radicals. Generally, ignition in HCCI engines is “triggered” by H₂O₂ decomposition [1] (Chaps. 15-18). However, a recent study [22] indicated that

reactive species produced by locally fuel-rich combustion can enhance main combustion for relatively late NVO fueling. In particular, hydrogen and acetylene were probed in [22]. Interestingly, that work also demonstrated that locally rich combustion may be caused by piston-top wetting and resulting pool fires. The latter are also investigated in the present study.

EXPERIMENTAL

Single-Cylinder Research Engine Set-Up

For the engine investigations a naturally aspirated direct-injection single-cylinder research engine is used. Injector and spark plug are placed in central cross position in the combustion chamber roof with the spark plug installed between the intake valves and the injector being installed between intake and exhaust valves. The engine is equipped with a fully variable electro-mechanical valve train, which provides high degrees of freedom regarding the desired valve timings. For the tests an outwards-opening hollow-cone piezo-actuated injector is used. More details of the commercially available injector can be found in [24, 25]. Tab. 1 provides an overview of the technical data of the test engine. The combustion-chamber geometry is outlined in Fig. 1. The matching all-metal engine is described in [8].

Table 1. Technical data of the research engine.

Displaced volume	499 cm ³
Stroke	90 mm
Bore	84 mm
Number of valves	4
Valve lift	8 mm
Compression ratio	12:1
Fuel injector	outwards-opening hollow-cone piezo (Continental Company)
Fuel pressure	100 bar

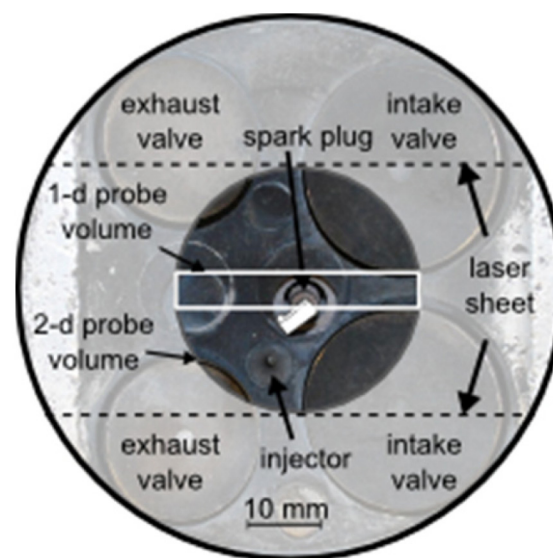


Figure 1. Combustion chamber and probe volume locations.

The cylinder, intake, and exhaust gas pressures are sampled with at least 1 °CA resolution over 300 consecutive engine cycles for the calculation of all indicated values. Higher resolution is used around top dead center. Pressure transducers and thermocouples are used for static pressure and temperature measurements. Oil and water conditioning systems allow steady-state operation and are set to 90 °C. The intake air is conditioned to 50 °C upstream from the throttle flap. The pressures upstream from the throttle flap and in the exhaust manifold are controlled to 1013 mbar during engine operation.

Combustion chamber recirculation (CCR) is chosen as the strategy for Controlled Auto-Ignition operation [26]. CCR is a type of internal exhaust gas recirculation (EGR) [8]. It is characterized by comparatively short valve opening events and therefore a long NVO period. The early closing of the exhaust valves enables high degrees of freedom in the calibration of different injection strategies.

The investigated operating conditions are specified in the following. A double fuel-injection event per engine cycle is used. The PI is triggered during the re-compression phase and the main injection during the intake stroke. The fuel-injection timing is varied in terms of start of PI (SOI₁) and duration of PI (DOI₁) while the sum of the durations of pilot and main injection is 520 µs leading to an approximately constant total fuel mass injected [27]. This is illustrated in Tab. 3. The fuel-mass values are measured by [27]. The “standard” operating conditions are shown in Tab. 2 and the investigated injection strategies in Tab. 3. The valve timing and thereby the EGR rate is only altered for a single fuel-injection timing. The amount of residual gas is calculated by a three-pressure analysis (TPA). The TPA is performed using measured intake and exhaust port pressure traces along with the measured in-cylinder pressure trace. The three pressure traces are imposed within a simplified engine model for the purpose of predicting quantities like species concentrations.

Table 2. Specifications of the selected operating conditions. All crank angles are given with regard to top dead center of main combustion (TDC).

Operating condition	“standard”
Engine speed	1500 rpm
EGR rate (by mass)	35 %
IMEP	~ 3 bar
Start of main injection (SOI ₂)	-260 °CA
Global relative air/fuel ratio (λ)	~ 1.3
Intake timing	-249 °CA – -170 °CA
Intake timing for reduced EGR	-254 °CA – -170 °CA
Exhaust timing	160 °CA – 286 °CA
Exhaust timing for reduced EGR	160 °CA – 291 °CA

The effect of DOI₁ variation on the auto-ignition phasing is discussed in the Main Combustion Phasing sub-section. All other operating conditions are constant. Spark assistance is not applied. Note also that the engine operation conditions are not optimized for ignition phasing, since it is the objective of this work to investigate the effects of the PI on the main combustion phasing. In order to start each experiment from a consistent baseline, after switching from spark ignition (SI) to GCAI operation the same dwell time is always applied before starting the data acquisition.

Table 3. Specifications of the selected durations of fuel injection (DOI₁/DOI₂). The crank angles of SOI₁ are given with regard to TDC. The operating condition with reduced EGR is marked with an asterisk.

SOI ₁ = 305 °CA (early SOI ₁)	SOI ₁ = 320 °CA (late SOI ₁)	Fuel Mass PI [mg]	Total Fuel Mass [mg]
90 µs/430µs	90 µs/430µs	0.25	11.56
120 µs/400 µs	120 µs/400 µs	1.39	11.89
160 µs/360 µs	160 µs/360 µs*	2.82	12.28
200 µs/320 µs	-	4.29	12.64
225 µs/295 µs	-	5.08	12.62

Optical Set-Up

The optical engine design is shown in Fig. 1. Optical access to the combustion chamber is provided by a glass ring and a window (diameter: 33 mm) in the piston. The entire optical setup is illustrated in Fig. 2. The LIF from formaldehyde is excited by using the radiation of a frequency-tripled Nd:YAG laser (355 nm) [18]. Its beam is formed to a horizontal sheet (40 × 3 mm²) and led through the glass ring, so that it covers the entire 2-d probe volume which is observed through the piston window. Thereby, measurements can be conducted at all CAs except in the ranges from -36 to +36 °CA with regard to both top-dead-centers, in which the laser beam is blocked by the piston.

The laser-pulse energy is about 50 mJ in the 2-d probe volume. Thus, the corresponding laser intensity is of the order of 5 MW/cm². According to [28], the LIF signal increases linearly with increasing laser intensity up to about 2 GW/cm². Hence, the current measurements are conducted in the linear regime, so that shot-to-shot laser-pulse-energy variations (about 5 %) can be taken into account to get LIF signals which are independent of these fluctuations. For this purpose, the total energy of each laser pulse is recorded by a laser power meter, as shown in Fig. 2.

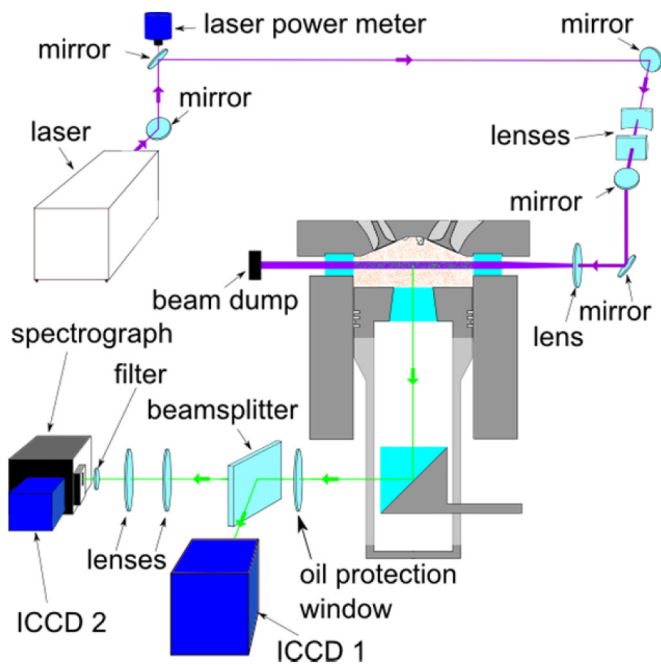


Figure 2. Optical set-up for LIF and chemiluminescence measurements.

The 2-d probe volume is imaged onto an intensified charge-coupled device (ICCD) via a 50% beam splitter and a lens. This camera is also equipped with an optical band-pass filter (405-505 nm) to get approximately interference-free formaldehyde-LIF images. The image intensifier of the ICCD is operated with a gate length of 5 μ s (unless otherwise noted).

Figure 2 also illustrates that the LIF transmitted by the beam splitter is imaged onto the entrance slit of an imaging spectrograph ($f_{\#} = 4$) via two spherical lenses ($f_1 = 500$ mm, $f_2 = 200$ mm) and a long-pass filter to suppress scattered laser radiation. The corresponding one-dimensional (1-d) probe volume is shown in Fig. 1. The spectrograph is equipped with a grating with 150 grooves per millimeter. Another ICCD camera is used in the exit plane of the spectrograph, in order to get both spatially and spectrally resolved images. The gate of the image intensifier of this camera is also 5 μ s. The chosen large detection-wavelength range yields information on possible interfering laser-stray light and in particular larger-PAHs LIF, which may appear at longer wavelengths compared to the peak of the formaldehyde-LIF [18]. For instance, the increased LIF signal at about 500 nm observed in formaldehyde measurements in an SI engine may be explained by PAHs [29].

Generally, LIF intensities depend on temperature and pressure variations as well as the quenching environment [18]. However, under non-combusting, relatively homogeneous conditions at -90 °CA in the compression stroke and at 225 °CA or 270 °CA in the exhaust stroke, the current measurements are basically affected by the temperature dependence of the LIF [13, 30]. Thus, it is taken into account in the data reduction, based on

the estimated, extrapolated data for engine conditions reported in [13]. It is assumed that the LIF intensity (per molecule), i.e. the quantum yield, increases with decreasing temperature by 0.2 %/K for the measurements at -90 °CA, where the temperature of the gas is about 540 K. In-cylinder temperatures are determined by pressure-trace analysis.

The current laser can only be operated with a repetition rate of 10 ± 2 Hz. Thus, it is not possible to fire the laser in each engine cycle at the used speed of 1500 rpm (12.5 Hz). However, the measurements at -90 °CA and 270 °CA are conducted in 2 of 3 engine cycles by operating the laser at 8.333 Hz, as shown in Fig. 3. Every second laser pulse is synchronized to the crank position of -90 °CA. The repetition rate of the laser is slightly adjusted for the image acquisition timings of the CAs 295 °CA, 180 °CA, and 225 °CA, so that synchrony with the engine is not lost.

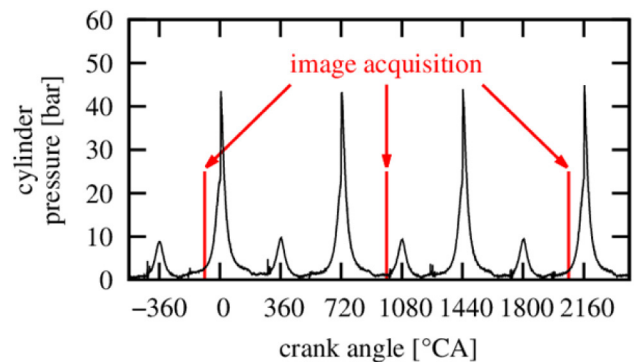


Figure 3. Image acquisition timing.

The described detection set-up is also used for natural-flame luminescence imaging. However, increased camera gate lengths in the range from 0.5 ms to 1 ms are employed to optimize signal throughput and temporal resolution. The extended gate length does not lead to substantial blurring of the chemiluminescence measurements. The effect of shot noise is discussed briefly in the "Chemiluminescence During Re-Compression" section.

RESULTS AND DISCUSSION

Formaldehyde Mixing

In order to demonstrate mixing of formaldehyde between recompression and compression, typical single-shot LIF images measured at -295 °CA, -180 °CA, and -90 °CA are shown in Fig. 4a), b), and c), respectively. Each image is measured in a different cycle. The field of view corresponds to Fig. 1, i.e., it is essentially limited by the piston window. However, the field of view is slightly larger than the piston window (diameter: 33 mm), when the piston is not close to the 2-d probe volume, as illustrated in Fig. 4b), c), d). In these measurements, the engine is operated with early SOI₁ (305 °CA) and DOI₁ = 120 μ s. The other operating conditions are given in Tab. 2.

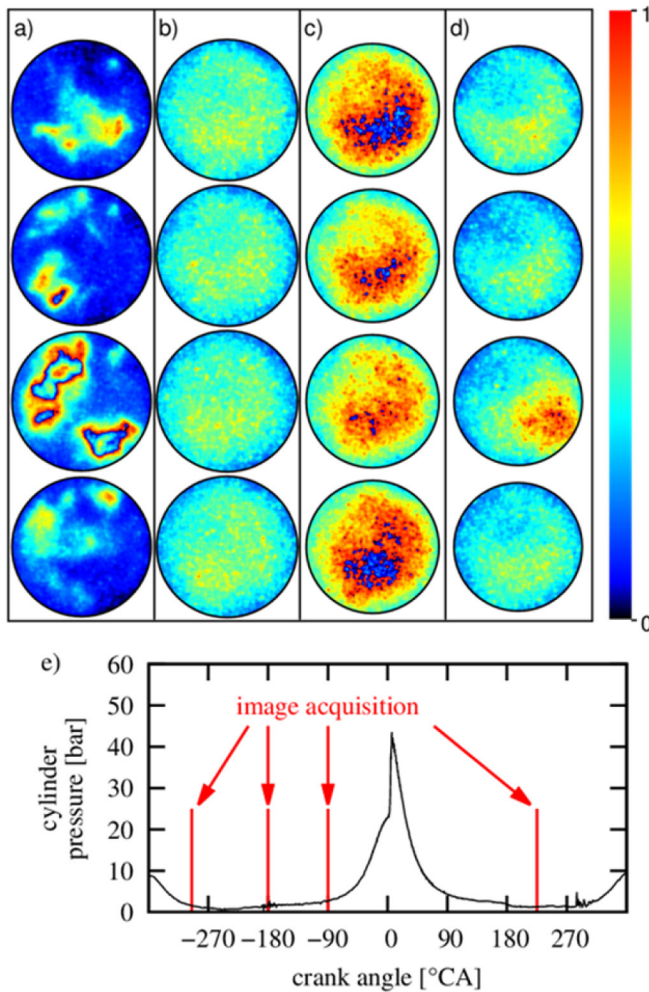


Figure 4. Typical single-shot formaldehyde-LIF images for $\text{DOI}_1 = 120 \mu\text{s}$ and early SOI1 (305 °CA) measured at -295 °CA in the NVO expansion stroke (a), at -180 °CA, i.e. bottom-dead center, (b), at -90 °CA during compression (c), and at 225 °CA in the exhaust stroke (d). Image intensities in b)/c)/d) are multiplied by a factor of two as compared to a). Note that the images in one row are not measured in the same cycle. Image acquisition timing (e).

Figure 4a) shows that highly inhomogeneous formaldehyde distributions are detected at -295 °CA, which fluctuate strongly from cycle to cycle. (It is demonstrated in the following subsection that the LIF signals are indeed basically caused by formaldehyde). Apparently, most of the measured formaldehyde is therefore produced by the combusting PI under these conditions. (Fig. 4a) shows that the detected overall formaldehyde density varies significantly from cycle to cycle. However, it can be assumed that this is partly caused by the limited field of view and the inhomogeneous distributions. This means that the overall formaldehyde density in the entire combustion chamber may be rather equal in each cycle.)

Compared to Fig. 4a), significantly lower LIF intensities are found at later CAs, as shown in Fig. 4b)/c) (note that the image intensities in Fig. 4b)/c)/d) are therefore multiplied by a factor of two, as compared to Fig. 4a), for better visibility). One major reason for this is the mixing with intake air. Obviously, rapid mixing also leads to substantially more homogeneous

formaldehyde distributions and low cycle-to-cycle variations in Fig. 4b)/c) compared to a). In fact, the inhomogeneity in the formaldehyde images in Fig. 4b)/c) is basically caused by limitations of the diagnostic. Firstly, the “spotty” structure of the images in Fig. 4b)/c) is dominated by quantum statistical noise (“shot noise”), i.e., it is induced by low signal intensities [18]. In order to reduce this source of error, the images in Fig. 4 are “smoothed” over ~0.3 mm in both directions. (The spatial resolution is essentially not affected, because it is limited by the vertical thickness of the laser-light sheet of 3 mm.) Secondly, decreasing signal intensities towards the edges of the images in Fig. 4b)/c) are basically attributed to the decreasing detection solid angle. Accordingly, these images are nearly rotationally symmetric. However, LIF intensities are noticeably reduced by window fouling in the upper left of each image.

In order to quantify the inhomogeneity in these images, they are averaged over 3 mm in both dimensions, so that shot noise is almost negligible (not shown). Thus, the spatial resolution equals 3 mm in all dimensions. The signal intensities in the central region hardly depend on other limitations of the diagnostic, such as the discussed attenuation at the edge or shot-to-shot variations of the laser-sheet profile. The latter are negligible. Thus, the single-shot images can be used to investigate the quasi-instantaneous inhomogeneity in the formaldehyde distribution. The relative standard deviation is found to be about 5 % and 4 % at 180 °CA and -90 °CA, respectively, when the central region with a diameter of 9 mm is considered. Therefore, these formaldehyde distributions are almost homogeneous. Consequently, the formaldehyde signals presented in the following subsections, which are averaged over the entire probe volumes of the 1-d and 2-d detectors, are assumed to be representative of all of the gas in the cylinder at these CAs.

The rapid mixing process illustrated in Fig. 4a) - c) appears to be roughly consistent with other comparable laser measurements [20, 31, 32, 33]. In the companion paper [20], local variations of fuel and residual gas content are reported to be only $\pm 10 \%$ at -40 °CA (for injection during the intake stroke). The air/residual gas mixing process was found to be essentially complete by -140 °CA in an HCCI engine, using planar tracer LIF [31]. Similarly, tracer LIF images showed that the air/residual gas mixture becomes almost homogeneous in the early compression stroke of a Controlled Auto-Ignition engine [32]. However, LIF signal variations of about 15 % were observed later in the compression (at -20 °CA), which were potentially caused by temperature fluctuations. (According to [31], the inhomogeneity in the temperature distribution increases in the late compression stroke.) Furthermore, cycle-to-cycle temperature variations of less than 10 % were measured by point-wise Coherent Anti-Stokes Raman Spectroscopy (CARS) at -90 °CA in an HCCI engine [33]. The corresponding variability in the air/residual gas content was found to be about 10 %.

Note also that the spatially and cycle-averaged LIF intensity, corresponding to the images in Fig. 4c), is by a factor of about 1.7 higher than in the case of Fig. 4b). This can be basically explained by both the doubled gas density and the increased temperature (by about 70 K) in the cylinder at the later CA in Fig. 4c). Thus, these measurements indicate that the LIF intensity per molecule is primarily affected by temperature variations, rather than variations of pressure, as noted in the Experimental section. According to the temperature dependence given there, the LIF intensity per molecule is expected to increase by about 20 % in the case of Fig. 4c) compared to b). (It is noteworthy that the measurements presented in Fig. 4b)/c) are conducted quasi-simultaneously, i.e., within one engine run, so that the LIF intensities can be precisely compared, in spite of varying window fouling.) This indicates that the formaldehyde detected at -90°CA is not formed by the igniting main injection (due to any possible fuel impurities), as assumed in the Introduction, but it seems to be primarily produced by the PI. (Thermal decomposition of formaldehyde occurs above $\sim 600\text{ K}$, i.e., after -90°CA [30]. According to pressure-trace analysis, the temperature is $\sim 540\text{ K}$ at -90°CA , as noted above.)

However, corresponding measurements conducted in the exhaust stroke (225°CA), which are depicted in Fig. 4d), show that there is a comparable amount of formaldehyde present before NVO. Due to the EGR, it can be concluded that the previous cycle contributes to the formaldehyde detected after NVO. Since combustion during NVO is relatively weak, as discussed below, it is assumed that the formaldehyde from the previous cycle is not completely consumed during NVO. (Note that there are small regions in the images detected shortly after NVO in Fig. 4a) where the formaldehyde signal vanishes. This can be explained by combustion of the formaldehyde from the previous cycle, which would otherwise be observed in the entire field of view.)

As shown in Fig. 4d), formaldehyde is found in the entire images. Thus, a certain fraction of the formaldehyde in the cylinder appears to be well mixed. However, inhomogeneities are also observed in some of the images acquired during the exhaust stroke, showing that some of the formaldehyde is formed shortly before the measurement. An example is depicted in the 3rd image in Fig. 4d). This may be caused by crevice mechanisms [34, 35]. Due to these inhomogeneities and the unknown fraction of formaldehyde consumption during NVO, the formaldehyde contribution to the following cycle cannot be precisely quantified.

Formaldehyde Identification

As noted in the Introduction section, formaldehyde-LIF may be affected by interfering LIF from PAHs. Chemiluminescence spectra presented below show that there is a fraction of fuel-rich combustion, which is particularly prone to PAH formation [18], during NVO. Thus, LIF spectra are considered to separate these potential contributions. A typical example is shown in Fig. 5. It is recorded at the same operating condition

that is used for the data depicted in Fig. 4 ($\text{SOI}_1 = 305^\circ\text{CA}$, $\text{DOI}_1 = 120\ \mu\text{s}$). The data acquisition CA is -90° , which is also applied for the compression-stroke measurements presented in the following. To reduce shot noise, the spectrum in Fig. 5 is averaged over 100 engine cycles and the entire 1-d probe volume. For comparison, a LIF spectrum from neat formaldehyde under ambient lab-air conditions is shown in Fig. 5. It is measured by the same detection set-up in the engine (without operation). In this case, the combustion-chamber is basically filled by a homogeneous air/formaldehyde mixture, which is produced by evaporating formalin (37 % formaldehyde, 10 % methanol, and 53 % water).

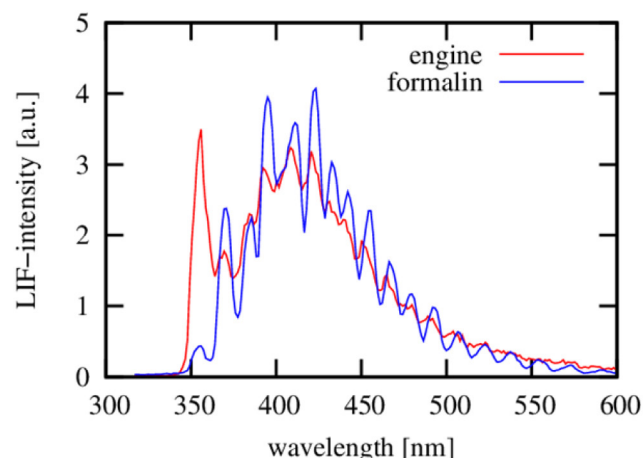


Figure 5. LIF spectrum measured at -90°CA and neat formaldehyde spectrum obtained by formalin evaporation.

Figure 5 shows that these spectra are very similar. (However, the relative intensity of the scattered laser radiation at 355 nm is significantly higher in the operating-engine spectrum, basically due to lower formaldehyde concentrations.) Comparable spectra were previously measured in cells, atmospheric flames, and engines, e.g. [28, 30, 29]. The well-known peaks of the formaldehyde emission spectrum, which are caused by transitions between different vibrational states [28], are resolved in both spectra in Fig. 5. However, these “lines” are broader and consequently the individual peaks are less distinct in the operating-engine spectrum. This is essentially consistent with previous results [28, 30, 29]. Presumably, this effect is caused by the increased temperature in the operating engine [30]. According to [28], it may also be attributed to broad, continuous, interfering emission from deposits on engine components. This would lead to a background similar to the one potentially induced by PAHs in the gas phase [18]. The emission from PAHs depends on the size of the aromatic structure [18] (Chap. 13). Small PAHs have peak emission wavelengths at about $(400 \pm 50)\text{ nm}$. If there was a noticeable contribution from small PAHs, the overall shape of the LIF spectra of neat formaldehyde and the gas in the engine would therefore be most likely different. For instance, the latter was indeed observed in [28]. However, the general shapes of the spectra in Fig. 5 are essentially equal (although the vibrational LIF peaks discussed above are less pronounced in the fired engine). Thus, LIF from small PAHs