

Two-Wavelength PLIF Diagnostic for Temperature and Composition

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ABSTRACT

Laser excitation wavelengths for two-line planar laser-induced fluorescence (PLIF) of 3-pentanone have been optimized for simultaneous imaging of temperature and composition under engine-relevant conditions. Validation of the diagnostic was performed in a motored optical IC engine seeded homogeneously with 3-pentanone. PLIF measurements of the uniform mixture during the compression stroke were used to measure the average temperature and to access the random uncertainty in the measurements. To determine the accuracy of the temperature measurements, experimental average temperatures were compared to values computed assuming isentropic compression and to the output of a tuned 1-D engine simulation. The comparison indicated that the absolute accuracy of the temperature measurements is better than $\pm 5\%$. Probability density functions (PDFs) calculated from the single-shot images were used to estimate the precision of the measurements. The PDFs determined from the images had standard deviations as low as ± 12 K for the temperature measurements. The diagnostic was demonstrated under conditions with controlled amounts of stratification induced by injection of cold unseeded fluid into the hot intake stream. Finally, the diagnostic was successfully tested during fired HCCI operation and was used to investigate the mixing of hot exhaust gas residuals with fresh intake charge.

INTRODUCTION

Temperature distributions have a strong influence on autoignition in both spark-ignition (SI) and homogeneous-charge compression-ignition (HCCI) engines. In SI engines one would like to avoid autoignition to keep knock from occurring. For HCCI combustion the autoignition process is the means by which combustion is initiated. HCCI combustion has the potential to have low NO_x and soot emissions while operating at high efficiency. Unfortunately, control of HCCI combustion is difficult and operation is typically only possible over a limited load and speed

range. Stratification of the in-cylinder temperature distribution has been suggested as a possible technique for altering the rate of heat release [1, 10, 14, 20]. Therefore, temperature stratification could possibly be used to exert control over the combustion phasing, enabling the expansion of HCCI to a larger range of conditions. Studying the influence of temperature stratification requires accurate measurement of the temperature distribution during the compression stroke. Temperature distribution measurements can also be used to diagnose problems with high NO_x emissions and knock in high compression ratio SI engines.

Temperature distributions under non-stratified conditions need to be characterized to compare with cases where stratification is induced. Measurement of the natural temperature variations present in-cylinder requires highly precise measurements of temperature differences as small as 10 K [18]. Many HCCI combustion strategies rely on substantial amounts of EGR present in-cylinder, so the measurement technique also needs to be insensitive to variations in composition. Additionally, since autoignition for gasoline-like fuels occurs at high pre-combustion temperatures, close to 1000 K, the technique should be capable of precise measurements at these conditions.

Planar laser-induced fluorescence (PLIF) is a diagnostic potentially capable of measuring the temperature distribution during compression with high precision and accuracy. Tracer-based PLIF has been used previously by Einecke et al. [5], Fujikawa et al. [7], Kakuho et al. [10], and others to measure in-cylinder temperature distributions. Two basic schemes were used by the investigators. The first involves capturing images from two excitation pulses (two-line excitation) usually applied in rapid succession [5, 7]. The temperature dependence of the ratio of the fluorescence signals is used to measure temperature. The second technique utilizes two tracers with different emission spectra and a single excitation wavelength [10]. If the correct tracers are chosen the fluorescence ratio can be calibrated to temperature.

In this work we chose the two-line excitation approach with 3-pentanone as the fluorescing molecule. Typically, the disadvantage of this approach is that it necessitates the use of two lasers for excitation and two cameras for collection of the fluorescence. We were able to eliminate the use of a second camera by utilizing an intensified CCD (ICCD) camera with an interline transfer array capable of acquiring two images in rapid succession. Use of a single camera eliminates the need to index the fields of view of two cameras to one another, and has the added benefit that it is no longer necessary to use a beam splitter; therefore, the collected signal for each image is increased by approximately a factor of two.

3-pentanone was chosen as the tracer since its fluorescence is only modestly affected by oxygen quenching, its absorption spectrum is easily accessible with high pulse energy laser systems, and its boiling point and transport properties closely match isooctane and n-heptane, typical model fuels. The modest dependence of 3-pentanone fluorescence signal on oxygen concentration is due primarily to rapid intersystem crossing [19]. When compared with aromatic tracers like toluene, 3-pentanone has much lower oxygen quenching, but also has a lower fluorescence quantum yield (FQY) and absorption cross section at room temperature. However, at temperatures near 1000 K 3-pentanone has a FQY for 308 nm excitation comparable to that of toluene using 266 nm excitation with N₂ as a bath gas. With air as a bath gas the FQY for 3-pentanone exceeds that of toluene [11]. In environments with substantial composition stratification, toluene is not a viable alternative due to the large influence of varying oxygen concentration on the fluorescence signal.

The absorption cross section for 3-pentanone has a usable level of absorption from 240 to 320 nm at room temperature which extends to a wavelength of approximately 350 nm at temperatures near 1000 K. Figure 1 shows the absorption cross section for 3-pentanone at temperatures of 300 and 1000 K. A number of high pulse energy laser wavelengths lie within the region of adequate absorption including krypton fluoride (KrF) excimer lasers (248 nm), the fourth harmonic of Nd:YAG lasers (266 nm), and xenon chloride (XeCl) excimer lasers (308 nm).

Although the boiling points of 3-pentanone (B.P. 375 K), isooctane (B.P. 372.4 K), and n-heptane (B.P. 371.5 K) [3] are closely matched, this does not guarantee the ideal co-evaporation of mixtures of these components due to non-ideal mixture effects. Whether preferential evaporation exists depends on the in-cylinder conditions at the time of the injection and the particulars of the injection such as the droplet size distribution [9]. Zhang and Sick [24] concluded that for typical engine conditions batch distillation of binary tracer mixtures is not significant. For most of the work in this paper the 3-pentanone is primarily premixed into the intake air upstream of the engine, and therefore, batch distillation of the droplets is not an issue.

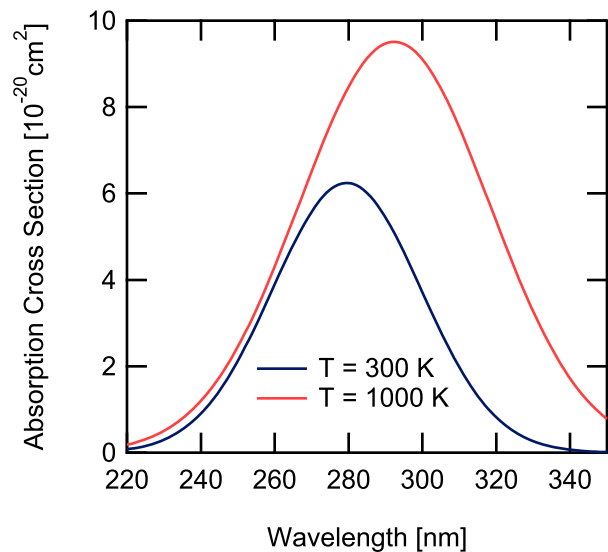


Figure 1: 3-pentanone absorption cross section at 300 and 1000 K calculated using the curve fit of Koch [11]. (Note: errors in the fit for $A(T)$, $\lambda_c(T)$, and $w(T)$, have been corrected and $A(T) = 4.77 \times 10^{-20} + 0.00487 \times 10^{-20}T$ [cm²], $\lambda_c(T) = 273.7 + 0.0186T$ [K] [nm], and $w(T) = 25.1 + 0.0101T$ [nm], where T is the temperature [K] (J. D. Koch, personal communication, December, 2007)).

Previous temperature measurements performed using two-line excitation of 3-pentanone utilized the easily accessible output of laser sources at 248 nm, 266 nm, and 308 nm [5, 12]. These laser sources are convenient, but are not necessarily at the optimal wavelengths for temperature measurements. The development of relatively accurate models for 3-pentanone fluorescence quantum yield and absorption cross section [11], and the recent measurements of 3-pentanone relative signal at simultaneously elevated temperature and pressure [2, 13, 23], allow for the estimation of temperature measurement precision as a function of laser wavelength and engine operating conditions.

In this paper we present the results of a simultaneous temperature/composition diagnostic utilizing two-line PLIF excitation of 3-pentanone which has been optimized for in-cylinder engine conditions. Optimal wavelengths of 277 and 308 nm were selected based on the results of a full uncertainty analysis. The diagnostic has been validated using in-cylinder measurements of temperature and composition under both motored and fired operation. Under motored operation measurements were made over a wide range of inlet conditions to test and calibrate 3-pentanone fluorescence models, and to access the accuracy and precision of the diagnostic. Motored measurements with controlled amounts of stratification served to validate the diagnostic under nonuniform conditions. A fired demonstration was successfully performed using

the engine using HCCI combustion. Under fired conditions, the nonuniform temperature distribution caused by exhaust gases mixing with fresh charge during the intake stroke was measured.

BACKGROUND THEORY

The PLIF signal in the linear excitation limit is given by

$$S_f = \frac{E}{h\nu} dV_c \left[\frac{\chi_{tr} P}{kT} \right] \sigma(T) \phi(T, P, \chi_i) \frac{\Omega}{4\pi} \eta_{opt} \quad (1)$$

where S_f is the number of photons incident per pixel at the detector or the photocathode of an intensified camera [photons/pixel], E is the local laser fluence [J/cm^2], h is Planck's constant [J s], c is the speed of light in vacuum [cm/s], ν is the frequency of the incident laser radiation [cm^{-1}], dV_c is the excited volume, i.e., volume corresponding to a pixel in the object plane [cm^3], χ_{tr} is the tracer mole fraction, P is the total pressure [MPa], k is the Boltzmann constant [J/K], T is the temperature [K], σ is the absorption cross section [cm^2], ϕ is the fluorescence quantum yield (FQY), Ω is the collection solid angle of the optics used for imaging the fluorescence, and η_{opt} is the optical transmission efficiency of optics and filters used in the imaging setup.

For temperature measurements we take the ratio of fluorescence signals from two excitation wavelengths. Using Equation 1 for the signal at each excitation wavelength and taking the ratio gives

$$\frac{S_{f2}}{S_{f1}} = C_{cal} \frac{E_2}{E_1} \frac{\sigma_2(T) \phi_2(T, P, \chi_i)}{\sigma_1(T) \phi_1(T, P, \chi_i)} \quad (2)$$

where the constant C_{cal} is determined by making PLIF calibration measurements at a known uniform temperature. The calibration constant C_{cal} is given by

$$C_{cal} = \frac{S_{f2}^{cal}}{S_{f1}^{cal}} \frac{E_1^{cal}}{E_2^{cal}} \frac{\sigma_1(T^{cal}) \phi_1(T^{cal}, P^{cal}, \chi_i^{cal})}{\sigma_2(T^{cal}) \phi_2(T^{cal}, P^{cal}, \chi_i^{cal})} \quad (3)$$

where S_{f1}^{cal} and S_{f2}^{cal} are the fluorescence signals from the calibration images, and the other parameters are as defined for Equation 1 but with values appropriate for the conditions of the calibration images. The calibration measurements can be used to correct for the laser sheet energy distribution if the shot-to-shot variation in laser energy is small, since then we can include the ratio of the laser fluences E_2/E_1 in the constant C_{cal} which cancels with the ratio E_2^{cal}/E_1^{cal} . Since the variables E_1 and E_2 represent the laser energy at each pixel location, the assumption that the shot-to-shot laser variation is small is only valid if laser sheet profile variation is small, and if shifts in the average profile are minimal during the time between acquiring the calibration and data images.

If we define a new parameter R_{pp} given by

$$R_{pp}(T, P, \chi_i) = \frac{\sigma_2(T) \phi_2(T, P, \chi_i)}{\sigma_1(T) \phi_1(T, P, \chi_i)} \quad (4)$$

and include the laser fluence ratio in the calibration constant

$$C_{cal} = \frac{S_{f2}^{cal}}{S_{f1}^{cal}} \frac{1}{R_{pp}^{cal}(T, P, \chi_i)} \quad (5)$$

we can further simplify the expression for the signal ratio (Equation 2) to give

$$\frac{1}{C_{cal}} \frac{S_{f2}}{S_{f1}} = R_{pp}(T, P, \chi_i). \quad (6)$$

Equation 6 indicates that by applying our calibration correction to the data we have measured the ratio of the photophysical parameters. This ratio is a function of temperature, pressure, and a slight function of species mole fractions. For 3-pentanone the species mole fraction dependence can be ignored over a large range of conditions. The ratio R_{pp} is a single-valued function of temperature and pressure for most wavelength combinations. We can calculate the ratio by calculating the absorption cross sections using the curve fit developed by Koch [11] (see note in Figure 1) and the FQY using the ketone FQY model originally developed for acetone by Thurber [21, 22] and adapted for 3-pentanone by Koch [11]. The details for the curve fit and model are described well in [11] and will not be discussed here for brevity. Having calculated R_{pp} as a function of temperature and pressure one can invert the function to get temperature as a function of R_{pp} and pressure, i.e., $T(p, R_{pp})$. Pressure is measured in-cylinder, and assumed uniform, and R_{pp} is determined from the ratio of the fluorescence signals and the calibration measurements.

In addition to the temperature, determined from the ratio of the fluorescence signals, we get a measurement of the local composition based on the signals at either of the two excitation wavelengths. The quantity we measure depends upon how the tracer is seeded and how we reduce the data. By seeding the tracer into the fuel we get a measurement of the local fuel mole fraction or concentration, alternatively, by seeding the intake air and the fuel, we measure the air-plus-fuel mole fraction. If we idealize the in-cylinder contents as consisting of three quantities, intake air, fuel, and exhaust gas residuals (EGR), and assume that all of the seeded tracer in the air and fuel is consumed during combustion [6, 17], then subtracting our measured air-plus-fuel mole fraction from 1 gives us a measure of the EGR mole fraction, i.e.,

$$\chi_{EGR} = 1 - (\chi_A + \chi_F) \quad (7)$$

where χ_{EGR} is the EGR mole fraction, χ_A is the air mole fraction, and χ_F is the fuel mole fraction. Deschamps and Baritaud [4] used a similar technique, in which they seeded the intake air with biacetyl, to estimate average EGR distributions. They termed the technique negative PLIF [4]. We keep their designation here, but abbreviate it as N-PLIF. For the rest of the paper the focus for the composition measurements is determination of the local EGR fraction using the N-PLIF technique.

For the fired engine measurements the intake air and fuel were seeded, and the N-PLIF technique was used to image EGR mole fraction. To calibrate the mole fraction measurements used in the N-PLIF equation, Equation 7, the fluorescence signal was divided by a calibration image taken under conditions of uniform temperature and tracer mole fraction. The calibration images at each of the respective wavelengths are the same as those used for the temperature measurements. Both S_f and S_f^{cal} are given by Equation 1; taking the ratio of these two signals and solving for $\chi_{tr}/\chi_{tr}^{cal}$ gives

$$\frac{\chi_{tr}}{\chi_{tr}^{cal}} = \frac{E^{cal}}{E} \frac{P^{cal}}{P} \frac{1/T^{cal}}{1/T} \frac{\sigma^{cal}(T)}{\sigma(T)} \frac{\phi^{cal}(T^{cal}, P^{cal}, \chi_i^{cal})}{\phi(T, P, \chi_i)} \frac{S_f}{S_f^{cal}} \quad (8)$$

The tracer mole fraction is directly related to the sum of the air plus fuel mole fraction by

$$\chi_{tr} = \chi_s(\chi_A + \chi_F) \quad (9)$$

where χ_s is the mole fraction of the tracer seeded into the air and the fuel. The ratio $\chi_{tr}/\chi_{tr}^{cal}$ is thus equal to

$$\frac{\chi_{tr}}{\chi_{tr}^{cal}} = \frac{\chi_s(\chi_A + \chi_F)}{\chi_s(\chi_A^{cal})} = \chi_A + \chi_F \quad (10)$$

where $\chi_A^{cal} = 1$ because the calibration images are taken under motored conditions with no fuel injection. Assuming the local laser energy remains constant between the calibration and data images, all of the variables in Equation 8 are known. To get the EGR mole fraction one just needs to apply the N-PLIF equation (Equation 7) and correct for temperature and pressure using the temperature PLIF measurement and the in-cylinder pressure measurement, respectively.

EXPERIMENTAL METHODS

OPTICAL ENGINE A single-cylinder optical engine was used for the engine measurements. The engine is mounted on a Labeco CLR engine base and speed is controlled via a 50 hp GE electric motor. The in-cylinder geometry consists of a pentroof head and flat piston. The head is a GM prototype with three valves, a centrally located injector, and 2 spark plugs. Figure 2 shows the relative locations of the injector and valves in the field of view used for the PLIF images. The Bowditch piston is outfitted with a 65 mm diameter window. Access to the in-cylinder components is provided by a drop-down liner system. The cylinder liner consists of two parts, a 25 mm tall fused silica ring and a water-cooled steel cylinder. Optical access for the laser sheet is provided by the fused silica ring, while imaging access is through the piston window. All measurements were performed at an engine speed of 1200 rpm. The crank angle convention used throughout is $-360 < \text{CAD} < 360$ where 0 crank-angle degrees (CAD) is chosen to be top dead center (TDC) of compression. The relevant engine specification are summarized in Table 1.

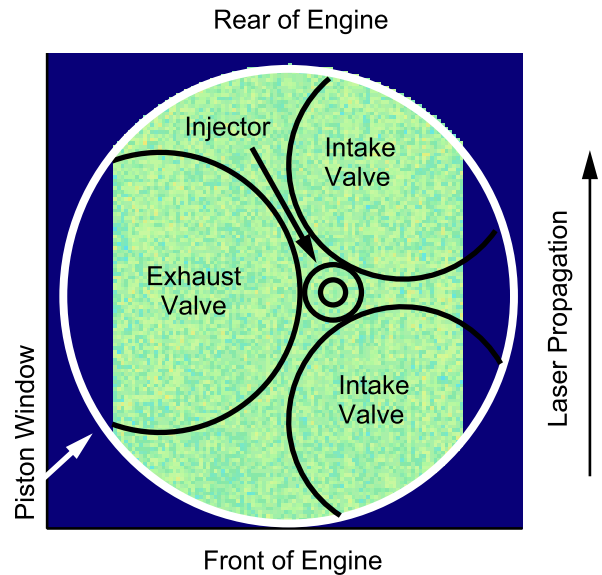


Figure 2: PLIF image with valve and injector locations superimposed.

Table 1: Engine specifications

Bore	92 mm
Stroke	95.25 mm
Compression ratio	11.5
IVO / IVC	-357 CAD / -212 CAD
EVO / EVC	212 CAD / 357 CAD
Engine speed	1200 rpm

PLIF SYSTEM The planar laser-induced fluorescence system consisted of two lasers, a Raman shifter, a single ICCD camera, and the ancillary optics. The laser wavelengths for two-line PLIF excitation were chosen based on the results of a full uncertainty analysis [16]. Parameter values used in the analysis applicable for the experimental setup used in this work are given in Table 2.

In the analysis, the precision of both temperature and composition measurements was estimated for the laser wavelengths listed in Table 3. The estimated profile variability listed in Table 3 is based on measurements and experience. The variability represents the variations in laser energy at a fixed location in the laser sheet. Estimates do not include the pulse-to-pulse total energy variation since this can be corrected for relatively easily by splitting off a portion of the beam and measuring it with a photodiode.

The range of in-cylinder temperatures and pressures considered are shown in Figure 3 with dashed lines indicating estimated temperatures and pressures for specific inlet conditions for the engine used in this study. The range covers pre-combustion temperatures and pressures typical for experiments in optical engines. The dashed red line corresponds to compression from a BDC temperature of 410 K and a pressure of 1.0 bar up to a temperature of

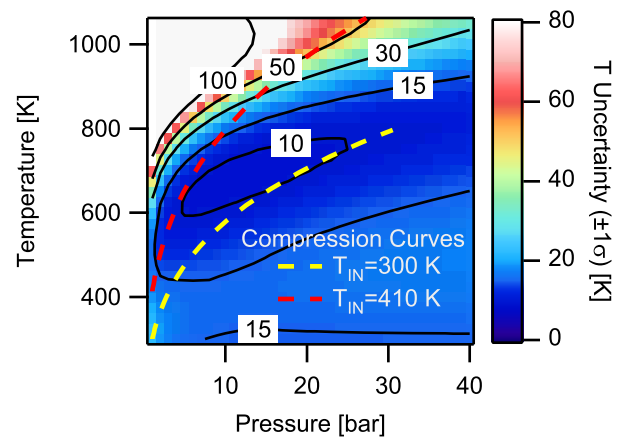
Table 2: Parameters used in the uncertainty calculations

Optics Parameter	Value
Lens f-number	1.4
Image magnification	0.21
Optics transmission	0.4
Camera Parameter	Value
Camera type	Intensified interline transfer ICCD
CCD array size	1024 x 1024
Pixel binning on chip	8 x 8
Binned pixels in linear extent	128
Binned pixel size in image plane	0.05 cm
Read noise	25 e^- per pixel
Photocathode QE	0.20
Total photoelectron gain	13
MCP noise factor	2.56
Laser Parameters	Value
Sheet width	50 mm
Laser energy	60 mJ if available else max from Table 3
Laser sheet profile stability	See table 3
Laser sheet thickness	0.5 mm
Other Parameters	Value
Tracer mole fraction in intake air	1000 ppm
EGR mole fraction	20%

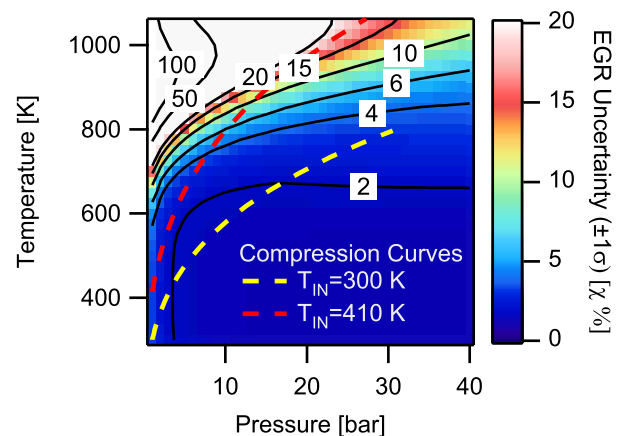
Table 3: Excitation wavelengths used in uncertainty analysis

λ [nm]	Source	Energy [mJ]	Profile Var. ($\pm 1\sigma$) [%]
248	KrF excimer laser	150-700	1
266	Nd:YAG 4 th harm.	100-200	5
277	KrF 1st Stokes in H ₂	15-80	2
289	266 1st Stokes in D ₂	20-40	5
308	XeCl excimer laser	200-500	1
332	XeCl 1st Stokes in N ₂	10-25	2
351	XeF excimer laser	200-400	1

1050 K. Temperatures higher than 1050 K are not considered since rapid decomposition of the tracer at these temperatures is expected. The dashed yellow line corresponds to compression from a BDC temperature of 300 K and a pressure of 1.0 bar. The optimal wavelength combination for temperature and composition measurement over the range of engine conditions considered was determined to be 308 nm and 277 nm. Both of these wavelengths have low shot-to-shot laser profile variation and have higher absorption cross sections and FQYs at high temperatures than the more frequently used wavelengths of 248 and 266 nm. The estimated temperature and composition uncertainties for the chosen 308/277 nm combi-



(a) Temperature uncertainty 308/277 nm



(b) EGR uncertainty 308 nm (T from 308/277 nm)

Figure 3: (a) Temperature uncertainty estimated for 308/277 nm wavelength combination. (b) Uncertainty in EGR mole fraction measurement estimated for 308 nm excitation using temperature measured with 308/277 nm combination.

nation are shown in Figure 3.

A Lambda Physik COMpexPro 102 XeCl laser was used to generate the 308 nm laser pulses. The laser can provide greater than 200 mJ/pulse in a 20 ns FWHM pulse. The 277 nm wavelength was generated via Raman shifting of a 248 nm KrF excimer laser (Lambda Physik COMpex 102) to the first Stokes wavelength (277 nm) in H₂. The Raman shifter (Lambda Physik RS-75) was 90 cm long and used a 60 cm focal length lens to focus the beam and a 30 cm focal length lens to recollimate it at the exit. The shifter was filled to 25 bar total pressure with H₂ and the energy conversion efficiency to the first Stokes was approximately 10% when pumped with 400 mJ/pulse from the KrF excimer laser. The excimer lasers used in this work have low shot-to-shot energy variability ($\pm 1-2\%$ for 1 σ) and the shifted output variability ($\pm 2-3\%$ for 1 σ) was only slightly increased relative to the input.