Application of multi-parameter laser-induced fluorescence with amine/ketone tracer mixtures in DISI engines

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Abstract Modern IC engine concepts require complex mixture formation to achieve reduction of fuel consumption and pollutant emissions. For optimization of mixing processes, the temperature and air/fuel-ratio distribution have to be controlled precisely. The simultaneous detection of these parameters allows for an improved understanding of mixture formation and its effects on combustion. Planar laser-induced fluorescence is a suitable tool for visualizing air/fuel-ratio and temperature in combustion applications. In IC engines a tracer with known photo-physical properties is added to a non-fluorescent substitute fuel and excited by an UV-laser. The detected fluorescence signal depends on various quantities, including the desired parameters. This work presents a novel measurement technique for multi-parameter imaging in IC engines by applying laser-induced fluorescence with an amine/ketone tracer mixture. The selected tracers are triethylamine (TEA), which is a suitable tracer to visualize the air/fuel-ratio, and 3-pentanone, which is used for temperature determination in the injected fuel spray. These tracers were used before as single tracers for determination of one parameter each. In the present study both tracers are added to the non-fluorescent surrogate fuel isoctane. Prior to the application in an IC engine the applicability of the amine/ketone mixture has to be investigated with respect to the spectral properties, in order to ensure the separability of the fluorescence signals of both tracers. Furthermore, the spectral investigation can provide information on possible tracer interactions that influence measurement results. The spectral investigations were performed in a continuously scavenged calibration cell which can be operated at IC engine conditions. Additionally, the tracer mixture was applied in an optically accessible IC engine with exhaust gas recirculation. Averaged images and single shot images are presented showing the determined air/fuel-ratio and vapor temperature with the tracer mixture. The results shown demonstrate the applicability of the tracer mixture TEA/3-pentanone for simultaneous determination of air/fuel-ratio and vapor temperature.

1. Introduction

Mixture formation in modern IC engine concepts strongly influences fuel consumption and pollutant emissions. In direct injection spark ignition (DISI) engines with exhaust gas recirculation (EGR), mixture processes become more complex and have to be controlled precisely in terms of temperature, air concentration and air/fuel-ratio (AFR). An inhomogeneous fuel and temperature distribution may result in self-ignition and knocking in homogeneous supercharged engines due to local hot spots [2]. Reduction of NO can be achieved with exhaust gas recirculation leading to lower combustion temperatures. The determination of AFR, especially at the spark plug, is essential to judge ignitability. It is also necessary to identify local fuel-rich pockets, which lead to soot formation. For this reason, the measurement of temperature and AFR is of major importance. Planar laser-induced fluorescence (PLIF) has been used successfully to determine the parameters AFR, concentration or temperature [1, 4, 7, 8, 10]. Usually the addition of fluorescent tracers with known photo-physical properties is necessary for PLIF measurements in IC engines, which are selected by their sensitivity on the investigated parameter. Ketones, e.g. acetone and 3-pentanone, are mainly used for temperature distribution and air concentration measurements. Acetone is commonly seeded to the intake air [10], whereas 3-pentanone was successfully used for temperature determination in a fuel spray [12]. A suitable tracer for the determination of AFR is TEA, which provides strong oxygen sensitivity [1, 4]. Only few tracer mixtures have been investigated so far. Previous investigations of tracer mixtures were presented for studying mixing
in isothermal flows [6], for measurement of oxygen distributions [3, 7] or for temperature determination [5]. In a previous study [9] measurement of AFR, air concentration and temperature, which is important for a comprehensive description and understanding of mixture formation, has been demonstrated. For this purpose, two PLIF techniques have been combined for multi-parameter detection within the same combustion cycle. The tracer triethylamine (TEA), used for the visualization of AFR, and acetone, used for determination of air concentration and temperature, were combined and analyzed in respect to tracer interactions and their effects on the measurement results. The applicability of the novel measurement technique was proven by measurements in an IC engine with exhaust gas recirculation. The drawback of this tracer concept is that acetone is seeded to air and therefore no temperature and concentration fields can be measured at early mixing time points.

For this reason in this work another tracer mixture consisting of TEA and 3-pentanone for simultaneous determination of AFR and vapor temperature is investigated in terms of its applicability in IC engines. The focus of this work is on the applicability of temperature measurement in a fuel spray with the ketone 3-pentanone by using the two wavelength excitation PLIF when an additional tracer for AFR LIF, in this case TEA, is in the mixture. The tracer mixture is first investigated spectrally to ensure the spectral separability of the fluorescence signals of both tracers in a continuously scavenged high temperature calibration cell (HTC). Additionally, the spectra indicate possible tracer interactions which have to be considered when applying the tracer mixture. The tracer mixture is applied in a DISI engine with exhaust gas recirculation for multi-parameter imaging. AFR and vapor temperature distributions are shown to prove the applicability of the tracer mixture TEA/3-pentanone for multi-parameter detection.

2. Measurement principle

The fluorescence signal intensity depends on various parameters like e.g. temperature and mixture composition, which are investigated by the presented measurement technique. The fluorescence signal intensity is calculated by

\[ S_{fl} = \eta \cdot E \cdot \rho_{Tracer} \cdot \sigma(\lambda_{Exc}, T) \cdot \phi(\lambda_{Exc}, T, p, \chi_1). \]  

The fluorescence signal \( S_\theta \) is function of the optical efficiency factor \( \eta \), the laser pulse energy \( E \), the tracer partial density \( \rho_{Tracer} \), the absorption factor \( \sigma(\lambda_{Exc}, T) \), which depends on the excitation wavelength \( \lambda_{Exc} \) and on the temperature \( T \). Furthermore, the signal depends on the fluorescence quantum yield \( \phi(\lambda_{Exc}, T, p, \chi_1) \) which describes the probability of the molecule to emit fluorescence. It depends on the excitation wavelength \( \lambda_{Exc} \), the temperature \( T \), the pressure \( p \) and the bath gas composition \( \chi_1 \).

For exciting 3-pentanone two wavelengths are used, namely 248 nm provided by a KrF excimer laser and 308 nm provided by a XeCl excimer laser. Forming a signal ratio of the fluorescence signals emitted after excitation with different wavelengths eliminates the tracer partial density. The optical efficiency factor is eliminated by normalizing the signal to a reference. The measurements were performed in air, therefore, the bath gas composition is known. 3-pentanone is excited quasi-simultaneously (temporal delay is 5 \( \mu \)s) to ensure same flow conditions for both excitations. The obtained signal ratio is used to calculate temperature:

\[ S_R = \frac{S_{308}}{S_{248}} = \frac{E_{308} \cdot \sigma(308, T) \cdot \phi(308, T, p, Air)}{E_{248} \cdot \sigma(248, T) \cdot \phi(248, T, p, Air)}. \]  

By using calibration data, taken as a function of temperature and pressure, the temperature can be calculated by equation (2). The calibration data of 3-pentanone (see Löffler [11]) was investigated in a flow cell which can be operated at IC engine conditions. Further information about the calibration strategy can be found elsewhere [12, 13].

3. Spectral investigation
Before the tracer mixture is applied to an IC engine, spectral investigations were performed in a high temperature calibration cell (HTC) to study possible tracer interactions. The maximum operating conditions are 1000 K and 3 MPa. The cell is continuously scavenged with air and provides residence times of the tracer in the hot gas below 1 s. The amounts of tracer and carrier gas are controlled by using mass flow controllers and therefore the tracer partial density is known. 3-pentanone is excited by laser pulses with 248 nm and 308 nm wavelength which are formed to a light sheet. For spectral detection of the fluorescence signals an ACR Spectra Pro 275 monochromator with a grating of 150 g/mm and an Andor intensified CCD camera are used. The spectral investigation of the single tracers and the tracer mixture was performed at 423 K and 1 MPa in air. The ratio of the tracers in the mixture was set to 1:30 TEA/3-pentanone by volume. This ratio was selected to achieve best signal intensities of the tracers in the calibration cell. As TEA provides a relatively high fluorescence quantum yield a lower TEA concentration is required for suitable fluorescence signal intensity. Figure 1 shows the emission spectra of TEA and 3-pentanone as well as their mixture. To obtain additional information on possible tracer interactions the absorption spectra of both tracers are also included.

![Figure 1: Absorption and emission spectra of 3-pentanone [11] and TEA as well as the mixture of both tracers (excited with 248 nm) at 423 K and 1 MPa](image)

The emission spectra of TEA and 3-pentanone show only a small overlap between 310 nm and 360 nm. By choosing appropriate filters the signals can be easily separated. For this purpose, a filter set which is transparent from 270 nm to 310 nm is chosen for the detection of the TEA fluorescence signal. For 3-pentanone a 355 nm longpass filter is used for further measurements. The results show a significant overlap of the TEA emission spectrum and the 3-pentanone absorption spectrum. For this reason, tracer interaction is possible by the absorption of the TEA fluorescence signal by 3-pentanone. Such absorption would result in a lower fluorescence signal intensity of TEA and a higher intensity of the 3-pentanone fluorescence signal compared to the single tracer application. Thus, the signal intensities would be misinterpreted by using the tracer mixture without correction. Previous PLIF investigations using a TEA/ketone mixture showed that a tracer interaction due to the absorption of TEA emission could not be detected and therefore the tracer mixture could be applied without correction of interaction effects [9]. The ratio between amine and ketone is 1:100 by volume which is similar to the ratio used for the subsequent IC engine measurements. Signal changes in the 3-pentanone fluorescence due to tracer interactions are less significant than other error sources. It can be assumed that interactions are small and do not have to be corrected.

### 4. IC engine measurements

The tracer mixture was applied in an optical accessible DISI engine. The fuel injector is positioned on the side and the spark plug is located in the center. The tracer mixture consisting of
3-pentanone and TEA is added to the substitute fuel isoctane. TEA is seeded to the fuel with a concentration of 0.25 vol.-% and 3-pentanone with 20 vol.-%. The concentration is selected on basis of experience from successful PLIF measurements with the single tracers [1, 12]. In the transparent engine the influence of the added tracers on combustion are considered as described by Koch et al. [8]. A split-injection scheme is used for fuel metering corresponding to a catalyst heating point. During the first pulse two thirds of the overall fuel is injected starting at -280 °CA aTDC, and the second injection is finished at -60 °CA aTDC. The injection pressure is set to 6 MPa. The engine is operated at 0.35 MPa indicated mean effective pressure (IMEP) at 1200 min⁻¹.

For optical access to the combustion chamber, the IC engine is equipped with a fused silica glass ring. Two excimer lasers, providing 248 nm and 308 nm light pulses, are used for excitation of 3-pentanone (2-excitation-line LIF) whereas the same 248 nm pulse is used to excite TEA (1-excitation-line LIF). The pulse duration is 20 ns and the repetition rate is set to 2 Hz. The laser beams are formed to light sheets, which are overlapped by a dichroic mirror in the cylinder. The fluorescence signals of both tracers are detected perpendicularly with two ICCD-cameras (PCO, Dicam PRO). One camera is operated in a double-shutter mode to detect fluorescence of the quasi-simultaneous 2-excitation-line LIF signals of 3-pentanone. The selected filters avoid interfering fluorescence signal from the other tracer and scattering of laser light. The dichroic mirror is also used for previous separation of the fluorescence signals. Laser and camera are synchronized with the crank angle encoder by a pulse generator. To protect the glass ring from thermal overload, the operating time of the engine is limited to 4 minutes. A sketch of the engine setup is shown in Figure 2.

**Figure 2**: Optical setup at the DISI engine

The air/fuel-ratio (AFR) results are provided for correlation with evaporative cooling effects of the injected fuel/tracer-mixture. The presented crank angle ranges are -70 °CA aTDC to -30 °CA aTDC with a 5 °CA interval. For each crank angle 10 images were taken. Figure 3 shows averaged images of air/fuel-ratio and temperature during and after the second injection which is finished at -60 °CA aTDC.
The field of view is marked by the rectangle in each image. The areas beyond the region of interest (ROI) contain also scattering by the glass ring or piston which cannot be evaluated correctly. At -50 °C aTDC the piston is already moving into this field of view which is marked by the additional line. At -65 °C aTDC the injected spray is visible in the upper left corner of the field of view. In the AFR field the mixture in the spray region is very rich containing still a high amount of liquid fuel. Therefore, in the temperature distribution the spray is not visible but the spray front can be identified. The signal from liquid fuel cannot be evaluated. A naive evaluation would lead to temperatures below 293 K, which are set to zero and are visible as white areas. Some white marked regions stem from strong reflections, which could not be avoided at this stage of the measurement campaign.

Shortly after the end of injection (at -55 °C aTDC) rich mixtures are visible in the AFR distribution. Most probably, still some small amounts of liquid fuel disturb the 3-pentanone LIF-signal, which is only visible in the intensity ratio (i.e. temperature field). Evaluating the vaporized fuel, temperature is distinctly lower than in the right part of the image which is attributed to evaporative cooling. The spray is almost completely vaporized at -50 °C aTDC showing quantifiable evaporative cooling effects. The temperature in the left part of the image, in which mainly gaseous fuel is located, is about 350 K and comparatively low as compared to the temperature of about 450 K in the right part of the image. As expected, the regions of lowest temperature show also the richest mixture (AFR is about 0.5). The cooling of about 100 K is in the range of the measurement results of vapor temperatures conducted in an injection chamber by Trost et al. [12]. In single shot images the local temperature drop is even larger, which will be presented below.

Cyclic variations in mixture formation are responsible for events like misfire, knocking or high pollutant emissions. For this reason the single shot determination of AFR and temperature in the combustion chamber before ignition is very important. Therefore, single shot images of the mixing field are evaluated to characterize cyclic variations, see figure 4.
The result images show three shots at -50 °CA aTDC as the fuel/tracer-mixture is mainly evaporated at this point and only few regions in the ROI cannot be processed. The cyclic variations are mainly induced by the spray fluctuations. There are rich regions in the left part of the image which are not visible in the averaged fields. These could contribute to soot formation in some combustion cycles, which is critical for fulfillment of emission legislation. The fuel cloud also hits the piston and a strong temperature stratification is visible there. This could contribute to CO and UHC emissions as well. In the upper part close to the spark plug region the mixture is rich (AFR ≲ 0.5) whereas the right part of the cylinder shows a stoichiometric or slightly lean mixture. It can be concluded that the application of the tracer mixture TEA/3-pentanone is capable to resolve cyclic variations of AFR and vapor temperature in single shot measurements, which is valuable information for optimization of DISI-combustion concepts.

5. Conclusion

The efficiency of combustion in IC engines as well as pollutant formation is mainly determined by the mixture formation. Therefore, it is of particular importance to investigate air/fuel-ratio and temperatures before ignition. Tracer-based laser-induced fluorescence is a suitable tool for investigations of these parameters. For this purpose, this technique is extended for simultaneous determination of AFR and temperature by applying an amine/ketone tracer mixture. The tracer TEA was excited with one excitation wavelength to determine AFR. The simultaneous determination of the vapor temperature is performed by the 2-line-excitation LIF using 3-pentanone. The tracer mixture was investigated in terms of separability of the emission spectra and applied in an optically accessible IC engine. The tracer mixture was added to the fuel and a catalyst heating point with a split-injection scheme was investigated. Averaged and single shot images were shown and discussed regarding AFR and temperature distribution. The local average evaporative cooling is about 100 K, which is even larger in single shot images. The corresponding mixture field shows an average AFR of about 0.5. This novel measurement technique can also be applied to resolve cyclic variations which are sources of misfire of pollutant formation.

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6. References