Quantitative Analysis of Vapor Phase Structures in Transient Liquid Fuel Sprays

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ABSTRACT

Quantitative measurements of the fuel vapor phase of a heavy-duty Diesel engine common-rail injection system are presented. The air/fuel ratio in the evaporating and combusting spray has been obtained along a line of 4.3 mm length, including the spray axis at a distance of 26.5 mm from the injector nozzle. Based on single-shot one-dimensional linear Raman scattering measurements, a technique has been developed, which provides quantitative information about the vapor phase in Diesel-type fuel sprays. By careful adaptation of the optical setup, the Raman signals can be clearly discriminated against present background influences. A polarization-selective optical detection setup has been used, and both polarization components of the optical signal have been detected separately and simultaneously. In a heatable high-pressure chamber, liquid n-hexadecane has been used as a test fuel, and vapor phase air/fuel ratios and local structures have been studied along a line. Measurements have been performed at different times after injection and for two different rail pressures. With this newly developed technique, the influence of rail pressure on the evaporation dynamics of Diesel-type sprays can be expressed quantitatively.

INTRODUCTION

Increased standards in air pollution legislation and growing customer demands on improved fuel economy have led to intensified efforts in the development of combustion engines with direct fuel injection. For DI Diesel engines as well as for spark-ignited Direct Injection Stratified Charge (DISC) engines, a detailed knowledge of the local fuel distribution and quantitative values for the fuel/air-mixture composition is indispensable.

A variety of non-intrusive optical, mostly laser-based measurement techniques has therefore been developed in the last years. Liquid fuel distributions can be determined by means of laser Mie scattering, delivering global spray parameters like spray cone angle and spray tip penetration [1-3], as well as highly resolved imaging of spray breakup mechanisms close to the nozzle [4]. Droplet sizes and velocities are measured by using LDA/PDA-systems [5]. The vapor phase has been observed by utilizing laser-induced fluorescence techniques (LIF) [6] and a simultaneous detection of both vapor phase and liquid phase fuel distributions is possible by using the laser-induced exciplex fluorescence (LIEF) [7,8]. However, there is a lack of quantitative information on the vapor-phase composition, since most of the fluorescence methods are subject to influences from local gas composition, temperature and pressure [7], and in many cases a correction of these influences is not possible or introduces severe measurement uncertainties.

Optical measurement techniques based on linear Raman scattering can deliver quantitative results in technical combustion systems. Information on local gas composition can be obtained without the ambiguities that result from the unknown physical conditions in the measurement region and the influences from residual gases, which have to be taken into account when LIF-measurements are performed [7]. The effects of temperature and pressure on the Raman signal are considerably smaller and well known so that quantitative results can be obtained by performing a relatively simple calibration measurement. Consequently, the Raman scattering technique has been applied to technical combustion systems in the past [9-11]. However its signal weakness has very often prevented the use of the technique in heterogeneous combustion systems, where elastic scattering from fuel droplets can be up to twenty orders of magnitude stronger than the Raman signals [12], and therefore, only a few results have been published previously [13]. Especially in the dense spray regions close to the nozzle and on the spray axis, strong interferences from Mie scattering occur. Here, the wavelength-selective elements which are used in the optical detection path do not offer sufficient stray light rejection so that the Raman signals are completely covered by a very strong background. Furthermore, emissions from LIF, laser-induced incandescence (LII) or chemiluminescence can cause strong interferences which either lead to systematic errors or even make Raman measurements impossible [14].

In our work, we demonstrate the feasibility of one-dimensionally resolved Raman measurements, even on the spray axis of a common-rail heavy-duty Diesel injection system. In order to sufficiently suppress background
emissions, and to clearly separate the Raman signals, the measurement system has been carefully adapted to the measurement problem, and a number of factors have been considered when the optical system has been designed.

EXPERIMENTAL

Principle of Measurement
In order to determine the air/fuel ratio in an evaporating spray under Diesel-engine conditions, the density of air and fuel vapor has been measured separately and simultaneously by linear Raman scattering. We have decided to determine the density of air from the nitrogen rather than from the oxygen Raman emissions, since the density of nitrogen in air is about a factor of four higher [13]. This yields an increased S/N-ratio and since the spectral position of the nitrogen line is more separated from the elastically scattered signals, the interferences from stray light, e.g., by liquid fuel droplets are considerably reduced. The disadvantage of this procedure is that quantitative measurements can only be performed as long as no chemical reactions take place which change the nitrogen/oxygen ratio. However, since the technique has been mainly designed to study the evaporation and fuel/air mixing processes prior to ignition, this restriction is of minor importance.

As fuel, n-hexadecane has been used instead of Diesel, in order to receive quantitative results and to avoid fluorescence interferences from the fuel. This one-component fuel offers the advantage that the concentration of fuel vapor is directly proportional to detected Raman signal intensity, whereas with a multi-component fuel, fractional distillation effects would lead to ambiguities, even if the chemical composition of the fuel is well known: since the components of the fuel have scattering cross sections which differ from each other, but the spectral position of the detected Raman transition, namely the C-H stretching mode remains almost unchanged, the effective scattering cross section of the fuel vapor in the measurement volume changes during evaporation and thus, it is unknown. Therefore, quantitative measurements are only possible if one-component fuels are used or if the fuel consists of species that exhibit spectral properties which allow a separation of the different fuel components. We decided to use pure n-hexadecane, since with a boiling point of 560 K, this fuel represents very well the less volatile components within the Diesel boiling range. Also, many other physical and chemical properties, such as air consumption and dynamic viscosity match closely with the real fuel.

Finally, the detected intensity ratios of the spectrally integrated nitrogen and fuel Raman vibrational bands are converted into absolute values for the air/fuel-ratio λ by applying a constant calibration factor. The calibration measurements, which yield this relationship will be described below.

Optical Setup
The optical setup which has been used for the investigations is displayed schematically in Fig. 1. A frequency-tripled, pulsed Nd:YAG laser has been used for excitation, and a laser pulse energy of ~20 mJ at 355 nm emission wavelength was used to generate Raman signals. By choosing an excitation wavelength which is close to the visible range, interferences from fluorescence excitation of small molecules such as O₂ and OH, as well as from polycyclic aromatic hydrocarbons (PAH) can be avoided. At the same time, the signal to noise ratio (S/N ratio) compared to visible excitation sources remains high due to the λ⁻² dependence of the Raman scattering cross section. The maximum pulse intensity has been reduced by a factor of 5 by temporally stretching the laser pulse to a width of ~50 ns, which has benefits for the S/N ratio, since the threshold intensity for laser-induced gas breakdown is associated with a higher laser pulse energy [15]. The temporal pulsewidth also defines the temporal resolution of the measurement technique. By a set of spherical lenses, the laser beam is first expanded, and then collimated and focused into the measurement volume. This has been done in order to reduce the optical power densities on the optical accesses to the combustion chamber. A line-shaped focal region with a beam waist of ~7 μm defines the measurement volume in x- and z-direction. At right angle to the laser propagation direction (y-direction), a line extending from y = 0 mm to y = 4.3 mm is imaged onto the entrance slits of two spectrographs by a pair of spherical lenses with a magnification of 2. For the separation of the optical signal into horizontally and vertically polarized components, a Glan prism of birefringent calcite has been used. This prism offers a good polarization separation with a large acceptance angle and over a broad spectral range. Two imaging spectrographs (0.27 m, f/3.8) with 1200 l/mm gratings have been used for spectral resolution. Using imaging spectrographs in combination with 2-d detectors, it is possible to obtain a one-dimensional spatial resolution in addition to the spectral resolution. The slitwidth of the spectrographs has been set to 300 μm, which results in a high signal throughput at the expense of spectral resolution. However, the Raman lines of nitrogen and fuel have been clearly resolved. From Raman spectra of air, a spectral resolution of 0.8 nm has been determined. The signals were then detected by two 14-bit intensified slow-scan cameras (576 x 384 pixels) and stored on a computer for further data processing. The cameras have been operated in the gated mode with a gatewidth of 300 ns in order to

Fig. 1: Schematic of the experimental setup.
Fig. 2: Detected nitrogen/fuel intensity ratio $R$ as a function of the adjusted air/fuel ratio $\lambda$.

suppress continuous background signals, e.g. chemiluminescence. The camera pixels have been binned by a factor of 2 on the spectral axis and by a factor of 16 in spatial direction. Therefore, the recorded images had a format of 288 pixels on the spectral axis and 24 pixels on the spatial axis, and the resulting spatial resolution in y-direction was 200 $\mu$m. Since the spectographs possess short focal lengths and high f-numbers, their straylight rejection is rather poor.

Therefore, a holographic notch filter which suppresses elastically scattered light has been inserted into the optical detection path. In a spectral band of 9 nm width, centered around the laser wavelength of 355 nm, the filter has an optical density of 6. At longer wavelengths, the transmission rises from 60% at 390 nm up to 80% at 500 nm.

Calibration measurements

The Raman depolarization ratios of nitrogen and fuel are small, but not negligible, especially at elevated temperatures. Therefore, it is important to perform the calibration measurements with the same two-camera setup that is used later in the experiment. First, the relative spectral sensitivity curves of the two detection channels have been determined by placing a spectral calibration lamp with a known blackbody spectrum into the measurement volume. Then, a calibration of the optical detection system with respect to $\lambda$-values has been performed. In a heatable high-pressure chamber, different air/fuel ratios have been adjusted by injection of known amounts of fuel into the chamber by means of a microliter syringe and a septum. A series of calibration measurements for various air/fuel ratios has been performed and the obtained calibration curve is displayed in Fig. 2. The relationship between detected intensity ratio $R$ and the adjusted $\lambda$-value can be expressed by a simple equation:

$$ R = 0.626 \times \lambda $$

and thus, they are directly proportional. It has to be noted that the calibration measurements have been performed under the same pressure and temperature conditions as the measurements in the injection chamber. Thus, possible temperature and pressure influences on the shape and strength of the Raman emissions have been taken into account.

MEASUREMENTS IN A HIGH-PRESSURE INJECTION CHAMBER

The measurement technique has been applied to an evaporating spray in a high-pressure injection chamber. The chamber is supplied with pressurized air, which is heated to the desired temperature by a series of four electrical heating elements with a total power of 40 kW. The average flow rate of air is 0.1 m/s in the chamber, which is negligible compared to typical velocities in Diesel sprays and therefore, the air in the chamber can be regarded as quiescent atmosphere. The combustion chamber has a cylindrical shape with a diameter of 220 mm and a depth of 120 mm. In the chamber, air pressures of up to 60 bar and maximum temperatures of 800 K can be adjusted. The exciting laser is focused through the chamber by means of two rectangular optical accesses (115 x 60 mm$^2$) on the sides, and the optical signal is collected through a large circular window (Ø120 mm) on the front of the chamber. For our measurements, liquid fuel has been injected into quiescent air at a pressure of 45 bar and a temperature of 750 K. A heavy-duty Diesel common rail injection system has been studied. It is mounted to the back side of the chamber and consists of a fuel pump, a high-pressure radial piston pump, a fuel rail with pressure regulator and the injector, which was equipped with a 7-hole sac hole nozzle. Rail pressure and injection timing can be adjusted electronically. As it can be seen in Fig. 3, the measurement position is located 26.5 mm downstream of the injector nozzle. The detected line included the spray axis at $y = 0$ and extended in radial direction up to $y = 4.3$ mm. The air density in the chamber was 20.3 kg/m$^3$, and the injected liquid fuel volume was set to 40 mm$^3$ for all measurements. Measurements have been performed for 600 bar and 900 bar rail pressure for different times after start of injection. The resulting duration of the injection process was 790 $\mu$s for 600 bar rail pressure and 550 $\mu$s for 900 bar rail pressure. This corresponds to value of 25% of the maximum engine load.

Fig. 3: Location of the measured line relative to the spray. For comparison, a spray contour has been obtained by laser Mie scattering at 900 $\mu$s after start of injection. The circled position marks $y = 1.8$ mm.
Depending on the operating point of the injection system and the time after start of injection, different types of spectra have been obtained. At times less than 400 μs after start of injection, no significant number of spectra could be obtained, since liquid fuel droplets in the measurement volume lowered the threshold intensity for laser-induced gas breakdown [16], and intense broadband radiation from recombination of the generated ions is associated with this. These emissions cover the spectrum completely, and therefore no Raman spectra have been obtained at these times. Figs. 4 and 5 display spectra, from which air/fuel ratios have been determined. Type I is an undisturbed spectrum without significant background contributions. This type of spectra has been recorded predominantly in the early stages of the injection process. Figs. 4 (a) and (b) display the spectra which have been recorded by the cameras 1 and 2, respectively. The depolarized part of the C-H stretching mode of C_{16}H_{34} is clearly visible in Fig. 4 (b), whereas nitrogen shows only marginal contributions on camera 2, since its depolarization ratio is low [12]. The resulting difference spectrum is displayed in Fig. 4 (c).

The second type of spectra, from which air/fuel ratios have been determined, is shown in Figs. 5 (a) - (c). Formaldehyde is generated as a stable intermediate species during the cool flame reaction, which marks the beginning of the combustion reaction [17]. It is important to notice that in the cool flame, the temperature rise is less than 200 K, and less than 15% of the fuel has reacted [18]. Therefore, an evaluation of air/fuel ratios is still possible by the described procedure within the measurement uncertainties. However, as it can be seen from Fig. 5, the H_{2}CO fluorescence bands interfere both with the nitrogen and the fuel Raman lines, and therefore, a correction of the fluorescence background is inevitable. Fig 5 (c) demonstrates that these interferences can be removed efficiently by the polarization-resolved Raman technique. The difference spectrum shows no fluorescence contributions, and air/fuel ratios can be evaluated from these spectra correctly. These spectra have been recorded for up 1000 μs after start of injection.

Other types of spectra, which have been recorded, but not evaluated, included strong broadband PAH fluorescence during the main combustion reaction and spectra at the end of the combustion reaction which showed weak Raman signals of nitrogen and water as a combustion product.

RESULTS

For the two different rail pressures, 200 single-shot measurements have been performed at 400 μs, 600 μs, 800 μs and 1000 μs after start of injection. From these data, air/fuel ratios have been evaluated, and they are displayed as probability density functions (pdf), where the appearance frequency for a certain air/fuel ratio λ is plotted as a function of λ. It is interesting to note that the obtained pdf's did not show a dependency on the spatial location y, although one

![Figure 6](image-url): Gradients of the local air/fuel ratio in a single-shot image; dashed line: y = 0.94 mm, solid line: y = 3.20 mm.
half of the spray jet has been completely covered. This means that, on average, the air/fuel ratio is constant over the spray diameter which can be determined from the liquid fuel distribution. However, an investigation of the raw data reveals that in the single-shot profiles, gradients in the local stoichiometry are present. A typical example is given in Fig. 6, where a raw image is displayed together with two cross-sections which have been obtained at \( y = 0.94 \) mm and \( y = 3.20 \) mm. The air/fuel ratio close to spray axis is considerably lower than for larger distances from the axis. From this, it can be concluded that, although there are gradients of considerable strength present in the local air/fuel ratio, they are randomly distributed into positive and negative \( y \)-directions, and therefore they tend to cancel out on average.

Since no local structures were detectable in the pdf's, we will display our following results only for a single \( y \)-position. As a representative value, \( y = 1.8 \) mm has been selected. In Fig. 7 the obtained pdf's at 400 \( \mu s \) and 1000 \( \mu s \) are displayed for a rail pressure of 600 bar. A relatively broad distribution of \( \lambda \)-values, with a maximum on the fuel-rich side at \( \lambda = 0.5 \) has been determined. Over the observed temporal range, this distribution remains almost constant. The situation is quite different if the rail pressure is increased to 900 bar (Fig. 8): starting from very rich mixtures at \( \Delta t = 400 \mu s \), the distribution of \( \lambda \)-values shifts towards higher values at later times. From the obtained pdf's, weighted averages of the air/fuel ratio as a function of time have been calculated using

\[
\lambda(t) = \frac{\sum \rho_i(t) \lambda_i(t)^{-1}}{\sum \rho_i(t)},
\]

and the result is summarized in Fig. 9. The influence of the rail pressure on the temporal evaluation of the stoichiometry is clearly visible: while for 600 bar, fuel-rich mixtures are predominant until the combustion reaction starts, the averaged air/fuel ratio \( \lambda \) increases, if a higher rail pressure of 900 bar is adjusted. This effect is a clear indication of enhanced spray breakup and evaporation rates due to the higher rail pressure. Also, since at higher pressures, the same amount of fuel is injected in a shorter period of time, more time is remaining for the mixture formation process, until the flame front reaches the measurement location. Although only a small part of the spray region has been covered by the measurement technique, the results seem to indicate that for 600 bar, a large fraction of the spray volume contains rich mixtures, whereas for 900 bar, the average air/fuel ratio is clearly higher. However, even at 900 bar rail pressure, stoichiometric mixtures are detected only in a few cases, and it can be concluded that in the vapor phase, large areas of rich air/fuel mixtures are still present at the beginning of the combustion reaction. Two-dimensional measurements of the soot concentration during the combustion process by LII also agree very well with the data presented here [19].
CONCLUSIONS

The work presented in this paper clearly demonstrates the feasibility of Raman measurements under Diesel-engine conditions. By careful choice of the exciting laser wavelength (355 nm), major sources of interference, such as LII and LIF can be considerably reduced. By simultaneous detection of both polarization components of the optical signal, the Raman emissions can be clearly separated from the remaining unpolarized background. One-dimensionally resolved measurements of the local air/fuel-ratio of an evaporating n-hexadecane spray from a common-rail heavy-duty Diesel injection system have shown that, although local gradients are present in single-shot images, they are randomly distributed and tend to average out at the observed location. The positive influence of increased rail pressure can be found in larger air/fuel ratios at the beginning of the combustion reaction. However, even for 900 bar rail pressure, the mixtures remain fuel-rich in most cases. The results indicate that the presented measurement technique based on linear Raman scattering has been successfully adapted to the measurement problem and can be used for a comprehensive characterization of the vapor phase of Diesel-type sprays.

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REFERENCES

7. Fröba, A.P., Rabenstein, F., Münch, K.-U., and Leipertz, A., "Mixture of Triethylamine (TEA) and Benzene as a New Seeding Material for the Quantitative Two-}