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Development and Demonstration of 2D-LIF for Studies of Mixture Preparation in SI Engines

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Laser-induced fluorescence (LIF) has been developed for visualization of fuel distribution fields in an operating spark-ignition (SI) engine. Since the standard research fuel iso-octane, does not yield a useful LIF signal a fluorescent additive was used. None of the commonly used seeds were found adequate. A seed not commonly used in this context, 3-pentanone, $C_2H_5COC_2H_5$, was chosen due to favorable vaporization characteristics and fluorescent properties. Results from preparatory investigations in the actual engine environment are presented and related laboratory data are discussed. The two-dimensional LIF technique was applied to a spark-ignition engine and the fuel distribution at the ignition time was recorded. The resulting images were processed and converted into fuel/air equivalence ratio using an in situ calibration technique. The processed fuel distribution maps presented a noise level of ~ 10% and a systematic error not exceeding 0.03 fuel/air equivalence units. An increased combustion variability was observed when changing from a homogeneous to an inhomogeneous fuel/air mixture. Correlations of image data to the combustion development indicated that the increased cyclic variability could be largely explained by variations in the mean fuel concentration around the spark gap. The initial flame development therefore seems to be controlled by the average amount of fuel near the spark gap, whereas the actual distribution of the fuel within this volume is of less importance.

INTRODUCTION

During the last few years laser-induced fluorescence (LIF) has been developed for two-dimensional visualization of combustion parameters, [1, 2]. The LIF technique is species selective in both absorption and emission and has reported possibilities for detection of sub-ppm levels. The major drawback of the technique is that the signal is sensitive to collisional quenching, which is very difficult to quantify as it depends on the surrounding temperature, the density of all the collisional partners and their collisional cross-sections. In spite of this, LIF has been successfully applied to practical combustion devices, [3, 4]. Hence, 2D-LIF can be readily adapted for use in a spark-ignition engine to determine the fuel concentration field before ignition. The composition of conventional engine fuels, such as gasoline, is, however, not well-defined and may vary from batch to batch, making the fluorescent properties uncertain. The use of a single-component fuel, such as iso-octane, is therefore preferred. However, iso-octane does not yield laser-induced fluorescence and a seed must be added to it. The selection of seed is crucial to the success of the measurements. The seed should ideally have a high fluorescence quantum yield and laser-excited states that are insensitive to quenching by the principal collisional partners. Besides having reliable fluorescent properties, the seed should display similar vaporization characteristics as the fuel, so that the parent fuel can be marked adequately.

In this paper possible seeds to be added to iso-octane are evaluated and 3-pentanone, a seed which, at the time, never had been used in this context is investigated. Quite recently, however, 3-pentanone has been demonstrated for use as a seed in measurements of relative fuel concentration fields, mainly during the intake process, in a spark-ignition engine [5]. In the present work, the technique is applied to a spark-ignition engine where the LIF signal from the seed is captured immediately prior to ignition. The resulting fuel vapor distributions are subsequently processed and calibrated. Fi-

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nally, to further demonstrate the practical use of the technique, data are extracted from the images and correlated to the early combustion development.

APPARATUS

A typical arrangement for LIF imaging measurements in the SI engine is shown in Fig. 1. The laser was a Lambda Physik EMG 150 MSC operating with KrF around 248 nm. The laser radiation was formed into a vertical sheet of light by cylindrical fused silica lenses and was focused at the center of the combustion chamber in front of the spark plug electrodes. The final laser sheet was approximately 12 mm high by 0.2 mm thick and with an energy of ~ 100 mJ per pulse. The distance from the laser sheet to the *center* of the spark gap was approximately 3 mm. The fluorescence image was demagnified and focused by a conventional Nikon lens, $f = 50$ mm $f/1.2$, onto the image intensifier of a 14-bit, peltier-cooled, CCD camera system, ICCD-576S/RB-T, supplied by Princeton Instruments Inc. The active area of the CCD chip consisted of 384 × 576 pixels, each 25 × 25 μm^2. With a 2:1 demagnification and a resolution of the detection system measured to 2.5 pixels full width at half maximum [6], the resolution in the object plane was 125 × 125 μm^2. Colored glass filters were placed in front of the lens to reject undesired emission. The CCD camera and the laser were triggered by pulses from the engine crank shaft encoder. The laser firing and the camera exposure were set to one crank angle degree (CAD) before ignition, corresponding to 238 μs at an engine speed of 700 rpm. The gating time of the fluorescence detection system was fixed at 100 ns.

The experiments were in a single-cylinder spark-ignition engine based on a six-cylinder VOLVO TD 102 diesel engine. The top of one of the cylinders was extended by a spacer with three square fused silica windows (dimensions 38 × 38 mm). To maintain a reasonable compression ratio the original piston was extended. A disc-shaped combustion chamber geometry was used, in which a swirl ratio of 2.8 was generated in the standard diesel intake port. The engine was fueled on iso-octane (99.5% pure), seeded with a fluorescent species. When a homogenized fuel/air mixture was desired, the fuel was injected into the intake air more than 3 m from the intake port. To further improve the mixing, a 16-dm^3 mixing tank was

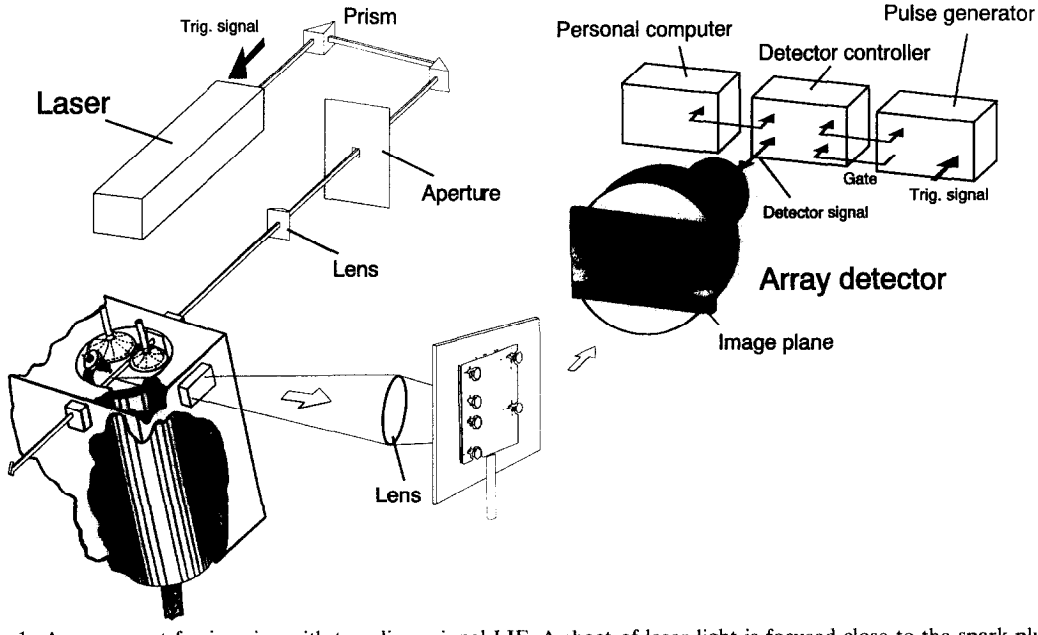

Fig. 1. Arrangement for imaging with two-dimensional LIF. A sheet of laser light is focused close to the spark plug in the engine combustion chamber. Emitted laser-induced fluorescence is collected at right angles and focused onto a two-dimensional array detector, controlled by a PC.

placed between this injection point and the engine. To produce inhomogeneities in the fuel/air mixture at the time of ignition, the liquid fuel was injected into the intake manifold close to the intake port, where it was allowed to vaporize and mix with the intake air. To minimize the amount of residual gas in the combustion chamber, the engine was run in a skip-fire mode, and the measurements made in the first clean cycle with combustion. The engine specifications are given in Table 1. The combustion process was followed during the pressure development, recorded by a piezoelectric transducer connected to a charge amplifier. The charge amplifier voltage output was sampled with a 16-bit A/D card and stored on a computer. To extract information on the early flame development a cycle-resolved heat release calculation was performed on the registered pressure data. For details of these measurements and calculations refer to Ref. 7.

CHOICE OF SEED

The difficulty of applying the LIF technique for studies of a non-fluorescing species is obvious. The investigated species has to be doped with a suitable substance which in turn can be detected by LIF. Fluorescence in engines has sometimes been measured without any knowledge of its origin. In diesel engines, fuel sprays have been visualized by UV excitation of presumed higher aromatic hydrocarbons, [8, 9]. In spark-ignition engines, the distribution of iso-octane has been studied after excitation by a KrF excimer laser [10, 11]. The origin of the fluorescence observed in these studies cannot be explained, as absolutely pure iso-octane does not fluoresce. This has been confirmed in our laboratory by spectroscopic investigations of iso-octane at STP conditions. In engine measurements, however, we have obtained weak LIF signals from iso-octane [12]. The unknown origin of this fluorescence makes signal interpretation hazardous; a volatile fluorescent impurity would be a poor marker of the fuel. The weakness of the signal further necessitates the use of a more strongly fluorescing additive. Such a seed was primarily sought among the well-known, previously used, flow field tracers such as acetaldehyde [13, 14], acetone [14], biacetyl [15, 16], and exciplexes, for example, TMPD (tetramethyl-*p*-phenylenediamine) [17, 18].

TABLE 1

Engine Specifications

Displaced volume	1600 cm^3
Bore	120.65 mm
Stroke	140 mm
Connection rod	260 mm
Compression ratio	7:1
Swirl number	~ 2.8
Engine speed	700 rpm
Skip-fire mode	3:7
Intake pressure	1 bar
Ignition time	14 CAD BTDC
Inlet valve opening (1 mm lift)	2 CAD BTDC
Inlet valve closing (1 mm lift)	42 CAD ABDC
Exhaust valve opening (1 mm lift)	39 CAD BBDC
Exhaust valve closing (1 mm lift)	4 CAD ATDC

In addition to possessing suitable fluorescent properties, the seed must also behave like the fuel. When mixing a liquid additive in a liquid fuel, the solubility and the vaporization characteristics should be considered as the main properties. Solubility in iso-octane is, however, not a problem for any of the seeds mentioned above. The importance of matching the vaporization characteristics, i.e., the boiling points, has been demonstrated in studies of multicomponent droplet gasification [19].

Acetaldehyde and acetone were rejected on the grounds of their excessive differences in vaporization characteristics compared with iso-octane [20]. The use of TMPD was ruled out as a result of the well-known strong quenching of its fluorescence by oxygen molecules, see Ref. 21. The fluorescence loss itself is not the limiting factor, but rather the introduction of an ambiguity in the interpretation of the resulting images. Biacetyl has been demonstrated as a possible marker of the iso-octane distribution field in a spark-ignition engine [22]. However, due to the incongruity between the vaporization of biacetyl and the vaporization of iso-octane, this seed was not regarded as optimum. One species with an almost perfect match of its rate of vaporization to that of iso-octane is 3-pentanone, also denoted as diethyl ketone, $C_2H_5COC_2H_5$ [20]. The absorption spectrum at room temperature,

ranging from approximately 220 to 320 nm with a maximum at 280 nm [23], is related to the forbidden $n \rightarrow \pi^*$ singlet-singlet transition [24]. The emission spectrum exhibits broadband emission from 330 to 600 nm with a maximum near 430 nm. The fluorescence quantum yield has been found to be essentially independent of excitation wavelength and nonradiative processes dominate over the radiative by 3 orders of magnitude [23]. To evaluate the potential of this compound as a seed, preliminary laboratory measurements were performed [25]. The signal intensity of 3-pentanone, at a maximum temperature of 200°C, was found to be almost independent of ambient air pressure in the range of 5–10 bar. Also the temperature dependence was found to be negligible over the range of 100° to 200°C at the relevant pressures.

The validity of adding 3-pentanone to iso-octane for LIF measurements was further tested in the actual experimental environment of the running engine. For these fundamental investigations the engine was fueled on a homogeneous charge created by fuel injection far from the engine. Excitation was at 248 nm where the fluorescence intensity was found to be directly proportional to the laser intensity over the range of 10 to 100 mJ. The amount of seed to be added to the iso-octane was determined by the fluorescence signal levels. The addition of 2% 3-pentanone to iso-octane, increased the fluorescence signal intensity by one order of magnitude. In the spark-ignition engine, the fluorescence intensity was monitored as a function of the injected amount of a mixture of 2% 3-pentanone in iso-octane. The LIF signal was found to be directly proportional to the injected amount of fuel up to the investigated equivalence ratio of 0.8. Absorption of the exciting laser light was very low. No decrease of the LIF signal could be detected over the field of view in the images of homogeneous fuel distributions. Using literature data [23] the laser beam absorption experienced after compression of a homogeneous, stoichiometric fuel/air mixture with 2% 3-pentanone in iso-octane was calculated to be less than 0.5% over the field of view [6].

Possible additive influences on the flame speed were investigated by comparing heat release data from the engine running on either pure iso-octane or on iso-octane with 2% or 5% added 3-pentanone, respectively. Conditions were not stable enough to allow detailed and definitive conclusions to be drawn, but any flame speed increase or decrease of more than approximately 20% would have been easily detectable. No such changes were observed. Nor have experimental investigations of flame speed changes by seed addition to iso-octane been found in the literature. Reference 26, however, reports a maximum increase in flame speed of 3% for lean/stoichiometric combustion, when 5% methyl ethyl ketone, $CH_3COC_2H_5$, closely related to 3-pentanone, was added to propane.

The decomposition of biacetyl in iso-octane has been measured to be approximately 11% per hour [22]. During the present experiments the decomposition of 3-pentanone in iso-octane was found to be much slower; the signal decreased by approximately 50% after 10 hours.

Based on all these considerations 2% (v/v) 3-pentanone was added to the iso-octane.

MEASUREMENTS AND RESULTS

The raw images of the fuel distributions had to be processed after acquisition to yield quantitative data. First, background data, obtained by motoring the engine without fuel injection, were subtracted from the original images. Second, the images were corrected for any nonuniformity over the profile of the laser sheet. The average spatial laser intensity distribution, which was extracted from images of homogeneous fuel distributions, was used for normalization. Variations in the total laser light energy would introduce ambiguities in the recorded fluorescence. Therefore, each image was normalized with the total laser intensity, which was monitored on a shot-to-shot basis by a power meter.

After the initial processing, the images contained the actual LIF signal from the doped fuel. This could be converted into fuel concentration by relating it to the signal obtained from a known fuel concentration field. Simultaneous cell measurements might yield a calibration factor as demonstrated in [27]. However, due to the difficulty of reproducing the

engine conditions of pressure, temperature and residence times an in situ calibration technique was developed. For every measurement sequence in the engine, measurements of homogeneous fuel distributions were made. By using conventional exhaust gas analysis [28], where the composition of the exhaust gas was related to the fuel/air equivalence ratio of the gas mixture fed to the engine, the LIF intensity in these homogeneous distributions could be assigned the corresponding value in terms of equivalence ratio. The images were accumulated and normalized by the laser mode cross-section and the total laser energy. The mean value in a section of the homogeneous image was used as a calibration value for all other images taken during the same measurement sequence. After this operation the images depicted the actual fuel/air equivalence ratio measured prior to ignition in the engine.

The fluorescence emitted from the laser sheet was found to be reflected on the shiny metal surface of the spark plug electrodes. Consequently, the area directly in front of the electrodes could not be reliably used for extracting data from the fuel distribution maps. The signal in the spark gap and away from the electrodes was not affected by the scattered fluorescence.

Processed measurements of fuel distributions from four consecutive engine cycles are displayed in Fig. 2. Strong inhomogeneities in the fuel distributions are undoubtedly present within each image, as well as from cycle-to-cycle. The color coding is shown beneath the images together with the corresponding calibration. From these distributions conclusions might be drawn on the importance of mixture preparation for combustion development.

STATISTICAL EVALUATION

To investigate whether the images could provide an explanation to the observed cyclic variations in the combustion development, a regression analysis was performed relating a combustion parameter to data extracted from the fuel distribution maps.

The crank angle position for a certain level of heat release was chosen as the combustion parameter. The level of heat release should be as low as possible to detect the flame at the earliest stage. The minimum level was limited, by measurement noise, to 0.5% of the total heat released, evaluated from the cylinder pressure by using an one-zone heat release model [7]. This level was used throughout the evaluations.

The objective was to see whether any correlation existed between image data and engine performance. Therefore, the first attempt should be as simple as possible and at the same time the extracted data should be physically relevant to the flame development. Calculation of the mean equivalence ratio *within a circular area* centered at the spark gap was identified as a reasonable point of attack. The radius of this circle was gradually increased and the average equivalence ratio for each radius was stored in a file. A measurement sequence typically consisted of 50 images. The above procedure was repeated for each of the 50 images. Naturally, before performing the data extraction, the images were processed and calibrated according to the procedures described earlier. After each measurement sequence two data files were finally to hand; one containing the position for 0.5% heat release for the 50 engine cycles, the other containing the average equivalence ratio around the spark gap as a function of radius for each of the 50 cycles.

The correlation of the mean equivalence ratio to the combustion parameter was expected to yield a second-order relationship, since laminar burning velocity is known to have an approximately second-order dependence on equivalence ratio [29]. The regression analysis was performed for every radius, recalling that the image data were computed within circular areas outwards from the spark gap. The correlation coefficient, which indicates how well the regression line explains the variance of the combustion parameter, was also computed for each radius. In Fig. 3a an example of the results from this analysis is displayed for images showing a high degree of inhomogeneity. The correlation coefficient as a function of radius is displayed together with the data scatter plot for the radius yielding the highest correlation coefficient. The maximum correlation is normally found for a radius of 5–10 mm

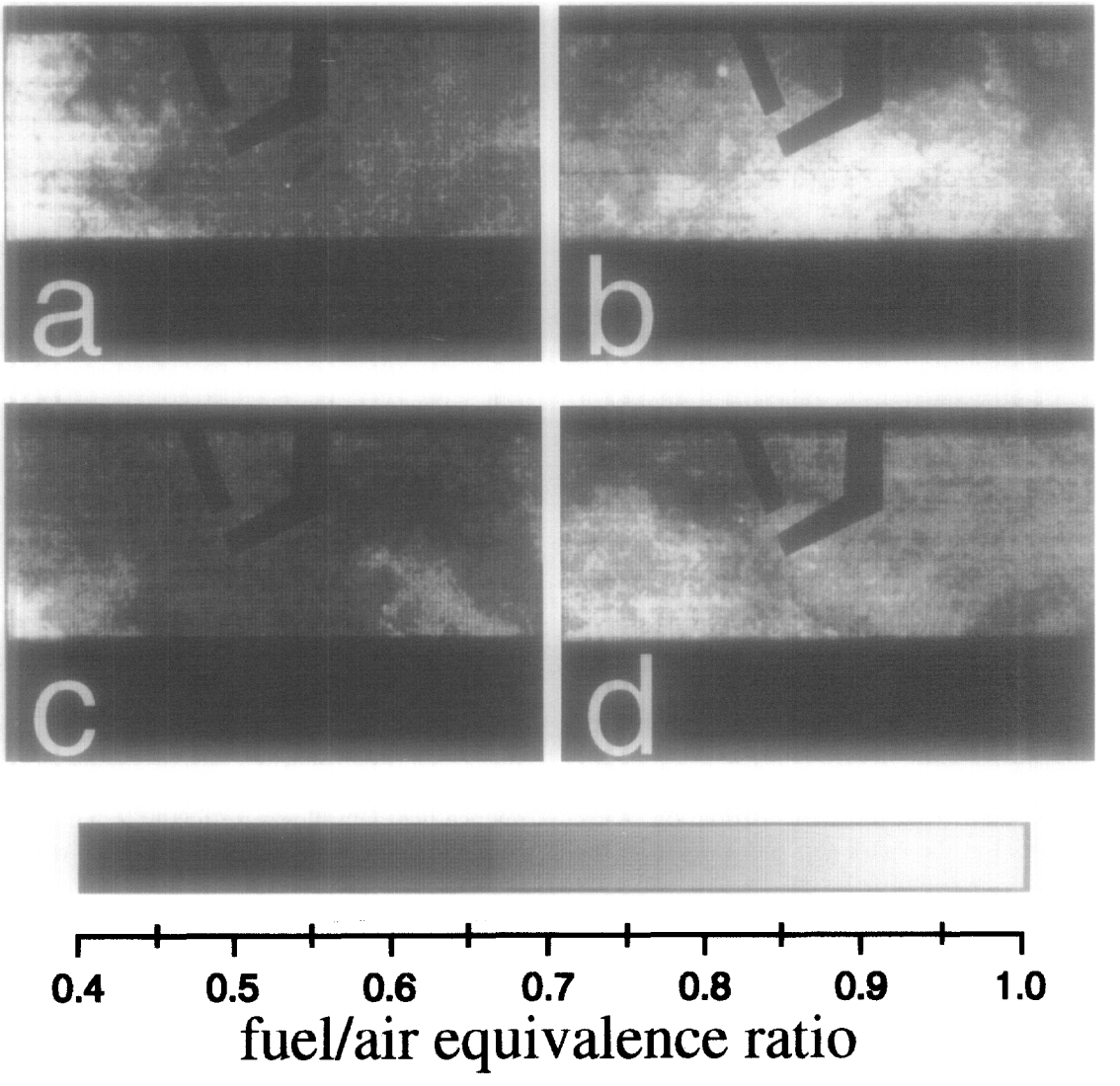

Fig. 2. Four consecutive, single-shot, fuel distributions measured 1 CAD before ignition in the engine shown with the color coding transferred into equivalence ratio. Field of view is 20 × 30 mm^2. Areas in front of spark-plug electrodes have been removed in the images.

from the spark gap. A strong dependence can be distinguished between the average equivalence ratio within a small volume in the vicinity of the spark gap and the early flame development. In Fig. 3b the corresponding results from fueling the engine on a homogeneous mixture are displayed. Obviously, the cyclic variation in average fuel concentration close to the spark plug is small and the resulting correlation coefficient decreases to a nonsignificant level.

A more detailed examination of the data has revealed that the major part of the increased combustion variability experienced for an inhomogeneous as compared with a homogeneous mixture, can be explained by variations in the average equivalence ratio contained within a small volume around the spark gap [6]. This nonreproducibility could be caused by instabilities in the fuel injection, but the total amount of heat released, which is proportional to the injected amount of fuel, only displayed variations of less than 1%. Therefore, the cyclic variations in mean fuel concentration at the spark gap prior to ignition were attributed to cyclic variations in the large scale flow history from injection to ignition.

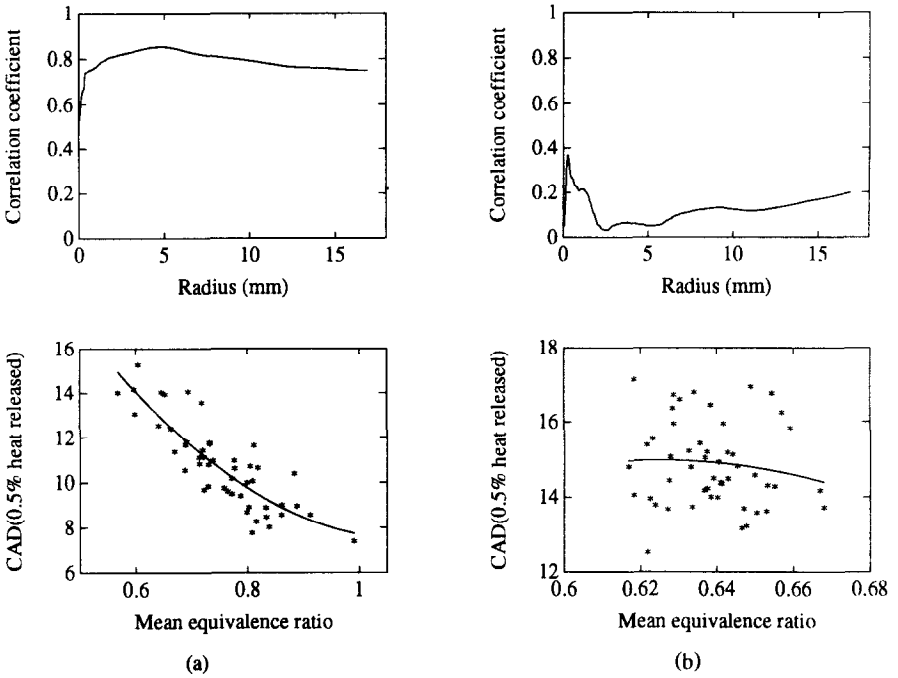

Fig. 3. (a) Inhomogeneous fuel/air mixture. Plot of correlation coefficient as a function of radial distance from spark gap (*top*). The data scatter plot (*bottom*) shows the position (CAD after ignition) where 0.5% of the total heat has been released versus the computed average fuel/air equivalence ratio. The plot is for the radius yielding the maximum correlation coefficient. Best second-order fit is indicated with a line. (b) Similar plots with a highly homogeneous mixture.

With a more advanced image evaluation technique, the images have the potential to give further insight into the importance of mixture preparation in internal combustion engines.

DISCUSSION

Assuming good agreement in physical properties between seed and fuel, the quality of the images and its influence on the results remain to be examined. The degree of pixel-to-pixel noise present in the images, Fig. 2, was determined by examining images of homogeneous distributions. After processing they presented standard deviations of ~ 10% at the rather high intensifier gain (9.0 of 10). The variations across the laser sheet from shot-to-shot have been measured as ~ 3% [6]. Hence, the major part of the image noise is created in the image intensifier. A larger fraction of seed, 5% (v/v), would allow a lower gain and, hence, reduce the intensifier noise. This pixel-to-pixel noise can also be reduced by post-processing, by convolution with a low-pass array, at the expense of a simultaneously reduced resolution, [30]. Noise due to lasermode instability can be eliminated by shot-to-shot referencing, where a second CCD camera records the beam profile for each image. Work in this field is currently in progress in our laboratory.

The extraction of single-shot data from an image, averaged in space as presented in Fig. 3, reduces the importance of the noise as it is averaged out in the process. Systematic errors, on the other hand, need to be evaluated since they affect the accuracy of the averaged data. No published data have been found on the dependence of fluorescence intensity on pressure and temperature at the present experimental conditions (15 bar/700 K). However, pressure variations are very small at the ignition time in the engine, less than 1%, and are unlikely to introduce uncertainties. Accordingly, assuming adiabatic compression, the temperature variations of the bulk gas should

be less than 0.3%. The pressure and temperature dependencies become important for mixing studies performed close to the walls, where boundary layers exhibit strong temperature gradients, and at different crank angles during the intake and compression phases, where both temperature and pressure change. The absorption of the laser beam over the field of view was shown to be low and can be omitted as a source of error. It was verified that no background level was building up in the engine during the measurements. The ratio of the fluorescence signal to the background level was always better than 15. An estimation based on the actual signal levels indicates that *if* the used background level were a worst-case, 50% underestimation of the real level, the resulting underestimation of the equivalence ratio would be less than 0.02. Errors could also be introduced through inaccuracy in the calibration value measured by the exhaust gas analysis equipment. However, the inaccuracy of this instrument should not exceed 0.01 in terms of equivalence ratio. Altogether, for spatially averaged data, a worst-case underestimation of 0.03 in equivalence units is possible.

SUMMARY

Examination of possible seeds for non-fluorescing iso-octane suggests the use of 3-pentanone, which has vaporization characteristics similar to those of iso-octane. Furthermore, this compound fulfills several basic prerequisites, adequate signal strength, linear dependence on concentration and laser intensity, low laser beam absorption, quenching independent of the principal collisional partners (O_2/N_2), low pressure and temperature dependencies. The technique has been applied to a spark-ignition engine where large spatial as well as cyclic inhomogeneities in fuel concentration were observed at the ignition time. After processing and subsequent in situ calibration into fuel/air equivalence ratio the images provided quantitative information. The image precision was limited to $\sim 10\%$ and the systematic errors were estimated to be less than 0.03 in equivalence ratio. A statistical evaluation using image data attributed the observed cyclic combustion variability to variations in the average fuel concentration close to the spark gap. Consequently, the initial flame development seems to be determined by the average amount of fuel near the spark gap, whereas the actual distribution of the fuel within this volume is of less importance.

The successful demonstration of the technique emphasizes its applicability. Two-dimensional, quantitative fuel distribution maps have the potential for providing new, tantalizing information relevant to, for example, the design of stratified-charge engines.

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COMMENTS

Michael Winter, United Technologies Research Center, USA. What is the effect of the presence of liquid-phase fuel on the accuracy of the technique? Does the presence of droplets directly effect the quantitative results?

Author's Reply. In the present work the fuel was prevaporized, which meant that no droplets were present during the experiments. We have therefore not investigated the effect of the injection process. Both tracer and fuel are well mixed in a gaseous form. In this case, the tracer follows correctly the fuel concentration flow in the cylinder. However, in a recent study of liquid film thickness [1] it has been shown that in a two-phase flow involving one major (iso-octane) and one minor (ketone) species, the evaporation rate of each component from the liquid phase is not the same as if they were separated. Repulsive molecular interactions cause a higher evaporation rate of the minor component then expected. This "distillation" phenomenon can be approximately compensated for by using a tracer with a lower vapor pressure curve than iso-octane, i.e., with a boiling point higher than iso-octane. In the same reference, 2-hexanone was found to be a practical and convenient tracer for iso-octane. With other fuels, other tracers must be sought.

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