

## ON THE QUANTITATIVE ANALYSIS OF VAPOUR FUEL FRACTIONS OF SPARK IGNITION DI SPRAYS BY USE OF LIEF

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### ABSTRACT

Laser Induced Exciplex Fluorescence is a diagnostic technique that generates spectrally separated fluorescence signals from liquid and vapour phases of a spray, providing temporal and spatial resolution of both species simultaneously. The technique is based on the model proposed by Melton [1] which requires two tracers to act as an electronic donor and acceptor pair forming an 'Excited complex' upon excitation by laser light at a certain wavelengths. The complex only reaches equilibrium in the liquid phase and produces a red-shifted fluorescence which allows characterisation of this phase. Measurements of the vapour phase are performed by tracking the fluorescence signal of the monomers. A mixture of triethylamine (TEA) and benzene in isooctane was chosen as the test fuel as it has very similar properties to the mid-boiling point fractions of gasoline and is expected to replicate its vaporisation efficiently. TEA and benzene form an exciplex complex upon excitation of a Nd:YAG utilising the fourth harmonic at wavelength 266nm. Two intensified CCD cameras were used to record the results as fluorescent images. Some preliminary experimental results of a transient spray from a gasoline direct injector are presented, with observations of the spatial distribution of vapour and liquid fractions. An insight of the limitations for quantitative results is presented with attempt to calibrate the vapour fractions of the spray using a two-phase characterisation methodology.

### INTRODUCTION

The visualization and quantification of fuel and air mixture distribution is of utmost importance in spark ignition engines. In gasoline direct injection (G-DI) engines working in stratified charge, small amounts of fuel are directed towards the spark plug so it is critical to know the location and concentration of the mixture to make sure there is a combustible mixture to ignite in every cycle.

All the G-DI combustion systems that have been proposed in the literature so far can be classified into three major groups depending on the primary strategy used to achieve stratification: spray-guided, wall-guided and air-guided systems [2, 3]. More recently, G-DI technology has adopted the 'spray-guided' concept as the preferred option as it significantly expands the speed/load range of stratified charge operation [3].

Quantifying fuel vapour fractions is particularly important for spray guided systems since the main mechanism used to achieve stratification is spray dynamics. Therefore, the main parameters to control stratification are: spray structure, injection timing and relative position between the injector and spark plug. For that reason it is critical to have a good understanding of the spray development, especially of vapour fractions, in order to contribute to design improvements for future G-DI engines.

Quantifying vapour concentrations in dense G-DI sprays is challenging since, for most contemporary optical diagnostic methods, the presence of liquid droplets can significantly affect the vapour signal. Some of the techniques available in the literature to quantify vapour fractions in multiphase systems are: Rayleigh scattering, Raman scattering, Coherent Anti Stokes Raman Spectroscopy (CARS), and Laser induced Fluorescence (LIF). CARS is rarely used in engines research due to its complexity, cost, and limitation to single point measurement. For Rayleigh scattering, the signal is strongly affected by Mie scattering of particles such as dust and liquid droplets. As Mie scattering is twenty or thirty orders of magnitude stronger than Rayleigh scattering, this technique shows little applicability for dense G-DI sprays [4]. Also, Raman scattering utilises very weak signals, and requires careful signal-to-noise consideration from background noise from sources such as: Rayleigh and Mie scattering from large particles, spurious scattering from optical windows and surfaces, flame luminescence and incandescence, fluorescence from excited molecules, and 'shot' and electrical noises of the detector. Again, the weakness of the signal, and the high presence of liquid droplets characteristic of G-DI dense sprays limits the applicability of this technique.

Laser induced fluorescence is widely used and potentially extremely useful for quantitative characterisation of transient flows fields, providing spatially and temporally resolved information in a desired plane. The wide applicability of LIF is due to the strength of the signal compared to Rayleigh and

Raman scattering, and because it provides spectral separation of different species. An extension of the LIF diagnostic technique known as Laser Induced Exciplex Fluorescence (LIEF) was proposed by Melton [1] to investigate the evaporation process of liquid sprays. This paper discusses the applicability of LIEF for spark ignition direct injection sprays with detailed consideration of the limitations for quantitative results.

## LIEF BACKGROUND

Laser induced exciplex fluorescence generates spectrally separated fluorescence signals from liquid and vapour phases of a spray, providing temporal and spatial resolution of both species simultaneously. The technique requires two tracers to act as an electronic donor and acceptor pair forming an 'Excited complex' upon excitation by laser light. The complex only reaches equilibrium in the liquid phase and produces a red-shifted fluorescence which allows characterisation of this phase. Measurements of the vapour phase are performed by tracking the fluorescence signal of the monomers.

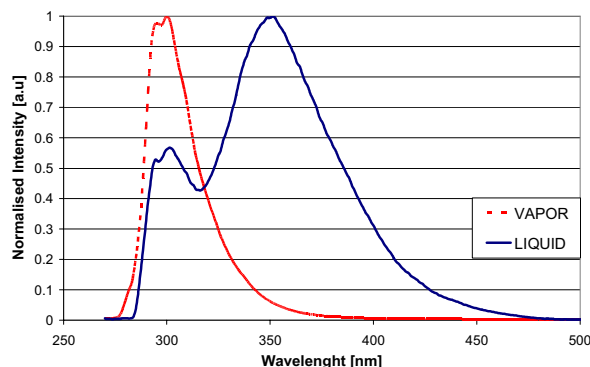
Melton proposed the use of N,N,N',N'- tetramethyl-p-phenyldiamine (TMPD) and naphthalene as exciplex fluorescent tracers for diesel applications [1,5]. Together, they form a loosely bound complex in the liquid phase that produces a red-shifted fluorescence band when excited with UV laser light. The monomer fluorescence of these tracers, used as a vapour marker, peaks around 380 nm when excited with laser light at 313 nm. On the other hand, the exciplex compound fluorescence peaks around 470 nm for the same excitation wavelength. Therefore, the liquid and vapour signals can be spectrally separated with careful selection of filters.

The TMPD-naphthalene exciplex tracer combination has been used extensively in the literature for diesel applications [5-9]. However, the boiling point of these organic compounds is very high compared to the mid-boiling points of gasoline. For this reason, these tracers are poorly suited for characterisation of gasoline sprays, as they would not replicate evaporation effectively. Melton [10] proposes using a combination of triethylamine (TEA) and fluorobenzene as fluorescence tracers suitable for gasoline applications. Although the co-evaporation of those tracers is excellent to replicate the evaporation process of gasoline, Melton reports considerable interference between the liquid and vapour signals.

Leipertz *et al* [11] proposes TEA and benzene as a suitable exciplex tracer combination for gasoline studies. These tracers have very similar properties to iso-octane and thus are excellent choices for simulating the evaporation characteristics of gasoline. They studied the combination of TEA and benzene very thoroughly publishing dependency of the tracers on temperature, partial pressure of nitrogen, partial pressure of oxygen, laser intensity, partial pressure of CO<sub>2</sub> and H<sub>2</sub>O, and air fuel ratio. They reported strong fluorescence quenching in oxygen presence, which is used to characterise air/fuel ratio. Again, considerable interference between vapour and liquid signal is reported, but the authors claim to achieve an unambiguous separation of the liquid phase fluorescence.

## TRACER SELECTION

The combination of triethylamine (TEA) and benzene proposed by Leipertz *et al* [11] was chosen from all the exciplex tracers available in the literature for its similar properties to the mid-boiling point fractions of gasoline. A solution of these tracers in iso-octane can be expected to replicate gasoline vaporisation effectively. The relative concentration of each component used throughout the experiments is: 2% of TEA, 2.9% benzene and 95.1% of iso-octane. (all percentages by weight).



*Figure 1:* Normalised fluorescence emission spectra of 2% TEA, and 2.9% Benzene in Iso-Octane. Note that the fluorescence of the vapour and liquid phases are not in the same scale.

Figure 1 shows the fluorescence spectra for the vapour and liquid phases of the exciplex solution, obtained with a Perkin Elmer LS50B Spectrometer. Please note that the normalised fluorescence of the liquid and vapour phases are not to scale. Although they are superimposed in this figure, no conclusions should be drawn regarding the relative intensity of the liquid and vapour signals. The fluorescence spectra shows a clear red-shift of the liquid fluorescence of about 50 nm. The vapour fluorescence peaks at 300 nm while the liquid signal peaks at 355 nm. At this wavelength, the vapour fluorescence is extremely low, allowing complete separation of the liquid phase. However, some fluorescence of the monomers is still visible in the liquid phase, and its contribution to the fluorescence of the vapour signal is probably significant. This crosstalk between the liquid and vapour signals makes quantitative measurements of vapour fractions very challenging. Therefore, a complete separation of the vapour phase emissions is only possible when the liquid phase emissions are recorded simultaneously, and some criteria is used in post-processing stages to identify or ignore liquid contributions in the vapour signal.

Leipertz *et al* [11] demonstrated strong quenching of the fluorescence signal by oxygen. For this reason, the experiments in this paper were carried out in a nitrogen atmosphere. For instance, in the presence of oxygen, high intensity could be the result of high fuel concentration, as well as low local quenching rates. For this reason, it was decided to eliminate the quenching variable for simplicity. Studying the fluorescence in oxygen-free nitrogen would allow a greater understanding of the crosstalk between the liquid and vapour signals.

## OPTICAL SETUP

Figure 2 shows a schematic of the optical set-up proposed for this technique in which two cameras are used simultaneously to record spectrally separated fluorescent images of liquid and vapour phases. A Nd:Yag laser utilising the fourth harmonic (266 nm) is used as the excitation light source. The laser beam is directed towards the rig and formed into an optically thin collimated laser sheet by means of various UV graded optics. The resulting laser sheet has a 35 mm height and a thickness of around 1mm throughout the interrogation area. In order to account for laser power fluctuations, an energy pulse monitor (EPM) was used to monitor and record the instantaneous laser power throughout the experiments. Even though the repetition rate of the laser is 10 Hz, the maximum repetition rate of the cameras is 5 Hz. Therefore images were acquired from several cycles in order to build a full fuel concentration map. For both cameras, the fluorescence signal was acquired through UV graded lenses onto a gated image intensifier linked to a HiSense Mk II CCD camera, with a resolution of 1344x1024 pixels.

For simultaneous detection of liquid and vapour signals each camera was equipped with different filters to optimise each signal at the peak of its fluorescent emission. A narrow band interference filter with centre wavelength at 300 nm, and minimum peak transmittance of 20%, was used to characterise the vapour fractions. For liquid separation, a combination of a long-pass cut-off filter and a narrow band filter was used to ensure no transmission below 350nm.

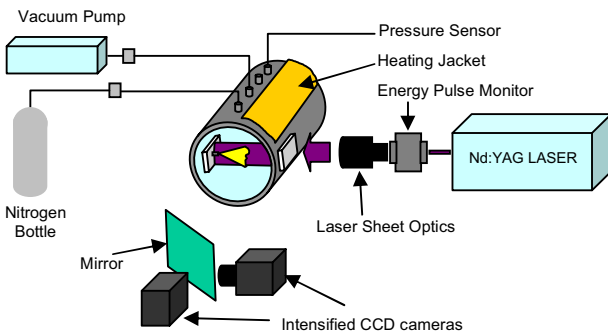


Figure 2: Schematic of optical Set-up

All experiments were carried out in an optical rig which allows independent accurate control of pressures up to 10 bar and temperatures up to 150 °C. A vacuum pump was used to evacuate the air of the rig between experiments. Then, the chamber could be re-filled with nitrogen to create a homogenous ‘oxygen free’ nitrogen atmosphere.

## CALIBRATION

In order to obtain quantitative results from LIEF techniques it is critical to establish the relationship between fuel concentration and the corresponding fluorescence intensity. Since the fluorescent signal is pressure and temperature dependent, it needs to be calibrated for each test condition. A preliminary study of temperature dependence of both

monomer and exciplex fluorescence is presented in figure 3. It shows that the intensity of the exciplex fluorescence decreases as temperature increases, in contrast to the intensity of the monomer fluorescence, which increases as temperature rises. For this reason, the experiments are conducted at 150 °C. At this temperature the contribution of the exciplex signal in the vapour detector is reduced significantly.

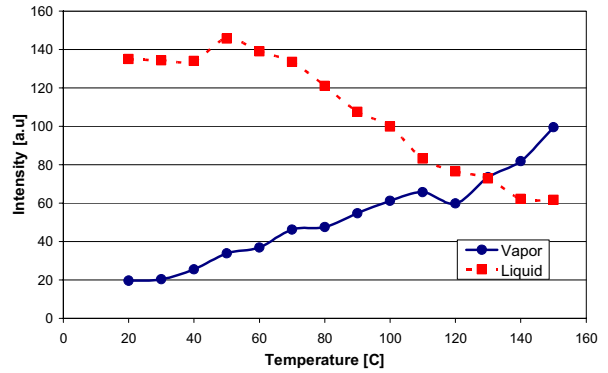


Figure 3: Relative fluorescence intensities of the liquid and vapor signals.

The calibration of the vapour signal is obtained by injecting a known mass of fuel into the rig after creating a vacuum and extracting all molecules of air. The fuel evaporates quickly as the test temperature is well above the boiling point of all components. Then, the chamber was filled with oxygen-free nitrogen until the test pressure of 1.4 bar (abs) was reached. The mixture was stirred with a fan to ensure homogeneity. Since the volume of the rig is known, the mean concentration of could be selected as desired. The monomer fluorescence intensity recorded for a range of Fuel-to-nitrogen ratios is presented in Figure 4. The figure shows that the fluorescence increases linearly with tracer concentration for these experimental conditions.

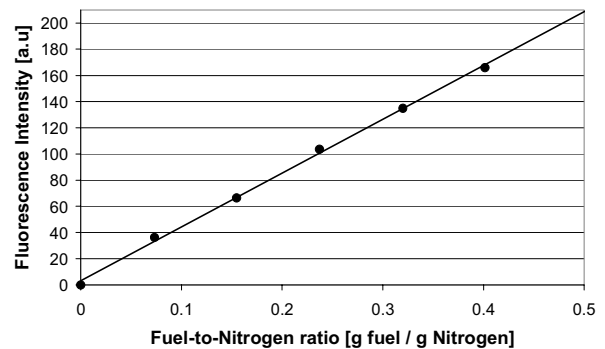


Figure 4: Variation of fluorescent intensity with Fuel-to-nitrogen ratio

## POST-PROCESSING

As mentioned previously, the main limitation for quantitative results when applying the LIEF technique is the considerable crosstalk of the liquid and vapour signals. However, research suggests that it is possible to achieve an unambiguous separation of the liquid phase fluorescence by careful filter selection [11]. Therefore, a separation of the vapour signal

could be possible if the contribution of liquid fractions in the vapour image is determined. This paper suggests using a binary image from the liquid signal to determine the location of liquid fractions in the spray. Later, the binary image can be used to identify liquid fluorescence on a vapour image acquired simultaneously. As a result, liquid fluorescence identified in the vapour image can be disregarded.

Before separating the fluorescence from the liquid and vapour phases, the images need to be corrected for systematic errors present in the imaging acquisition system and laser sheet irregularities. Consistent with Zhao *et al* [4] and Seitzman *et al* [12], the digital signal recorded for a single pixel of a CCD camera during LIF experiments is expressed as:

$$S_{pixel} = \left[ N_p \left( \frac{P_c}{P_o} \right) R_{total} + S_b + S_d \right] \quad (1)$$

where  $N_p$  is the number of fluorescence photons leaving the collection volume within the collection solid angle,  $P_c$  corresponds to the laser energy within the collection volume,  $P_o$  is the nominal laser energy,  $R_{total}$  the total responsivity of the detection system,  $S_d$  is the dark current signal, and  $S_b$  corresponds to the background signal associated with the laser induced process (e.g. Rayleigh and Mie scatterings, window fluorescence, and ambient light). Solving equation 1 for the fluorescence signal gives:

$$N_p = \left[ S_{pixel} - (S_b + S_d) \right] \cdot \left( \frac{P_o}{P_c} \right) \left( \frac{1}{R_{total}} \right) \quad (2)$$

Equation 2 suggests a suitable procedure for correcting systematic errors in the recorded image. First, an image combining the background and dark current signals ( $S_b+S_d$ ) is subtracted from the recorded image. This image can be obtained by removing the fluorescent particles from the system. Since the background varies from shot to shot, an average background is preferred to minimise this effect.

Then, the resulting image is then multiplied by a correction image to account for the spatial variations in the responsivity of the detector ( $R_{total}$ ). Finally, the new image is multiplied by another corrector factor to account for non-uniformities of the laser sheet ( $P_c/P_o$ ). Zhao *et al* [4] proposes recording the combined effect of the responsivity of the detector and the non-uniformities of the laser sheet by obtaining an image from a homogeneous mixture containing fluorescent particles ( $S_{uni}$ ) minus the background image ( $S_b+S_d$ ). Thus, the fluorescent signal of each pixel can be rewritten as:

$$N_p = \frac{\left[ S_{pixel} - (S_b + S_d) \right]}{\left[ S_{uni} - (S_b + S_d) \right]} \quad (3)$$

Once the errors in the acquisition system have been corrected as suggested by equation 3, the binary image obtained from the liquid fractions is used as a mask in the vapour image to disregard the fluorescence from liquid droplets. Finally, the corrected image of the vapour fluorescent signal can be calibrated for fluorescence concentration measurements by using the calibration curve presented in Figure 4.

## RESULTS AND DISCUSSION

Fuel concentration measurements by LIEF technique are presented in this section for a BOSCH C2-70 pressure swirl injector with a line pressure of 100 barG and injection duration of 1.5 ms. A solution of TEA, benzene and iso-octane is used throughout the experiments with relative concentration of: 2 %, 2.9 %, and 95.1 % respectively (all percentages by weight). The test was conducted in a nitrogen atmosphere with ambient conditions of 150 °C and 1.4 barA. Selected times after start of injection (ASOI) are presented with observations of the liquid and vapour distribution and fuel concentration for the vapour fractions. In order to attenuate systematic errors due to laser sheet power attenuation in dense sprays, only the first half of the images is processed.

Figure 5a shows an instantaneous liquid image recorded 1.25ms after start of injection (ASOI). The image has been randomly selected from 30 images acquired for each timebin. The image on the right corresponds to a binary image produced to identify liquid fractions in the spray. The sac volume can be clearly identified on both images which is characteristic of this type of injector. This injector is expected to produce a hollow cone, although liquid images of the spray show a full cone due to scattering.

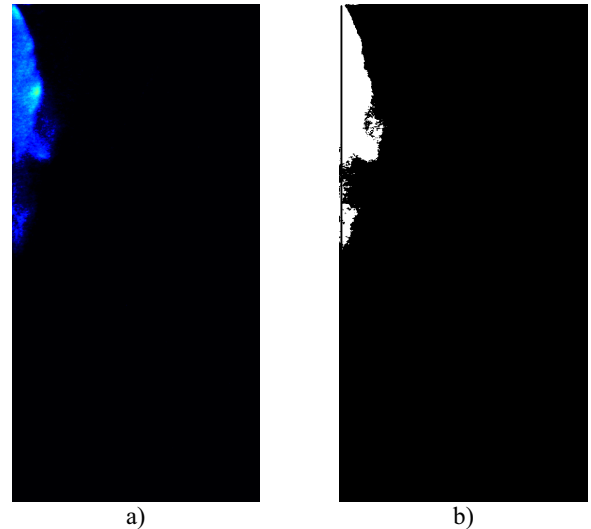
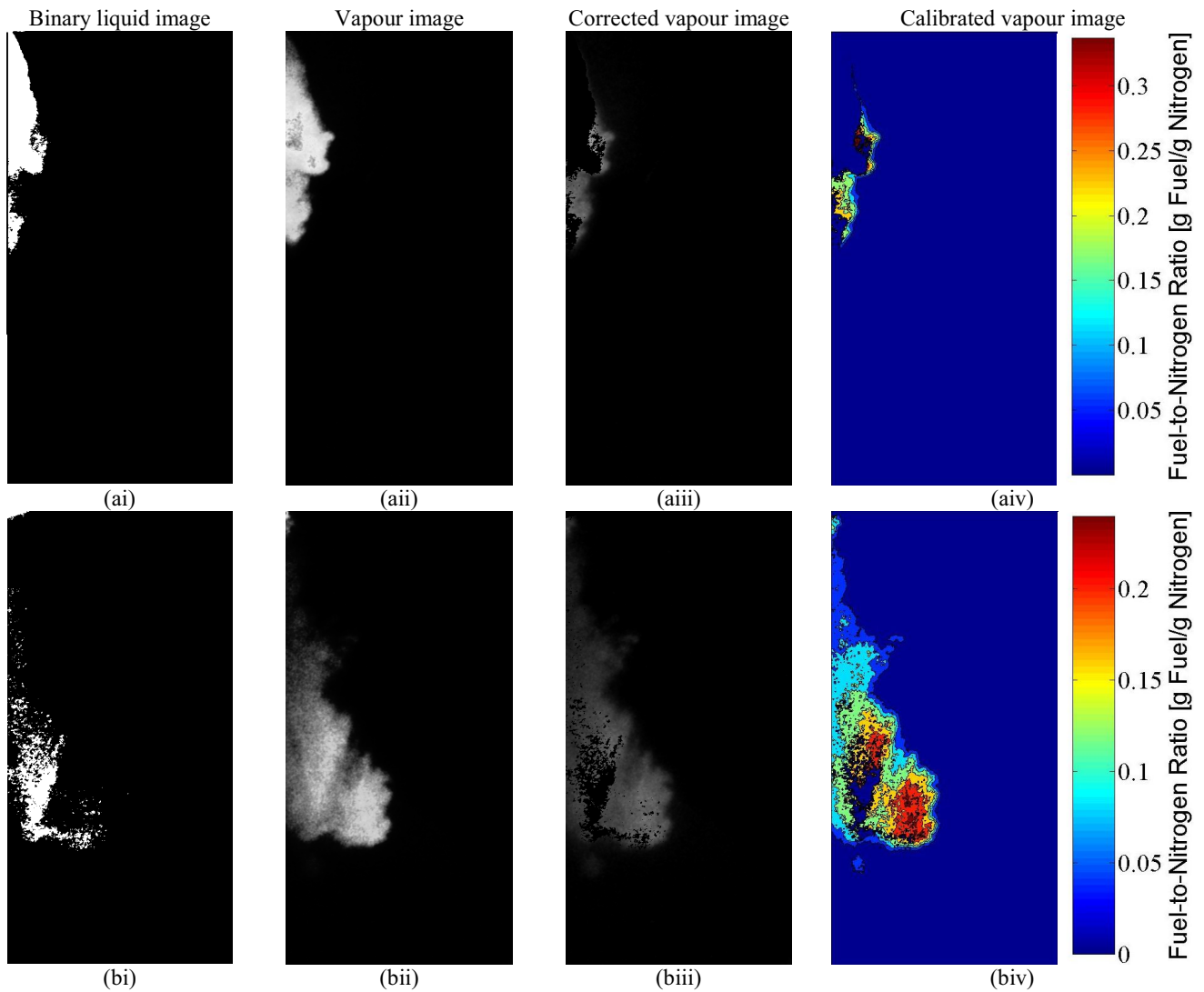


Figure 5: Liquid image 1.25ms ASOI. (a) image with background subtracted, (b) binary image)



**Figure 6:** Image processing sequence for two different timebins: a) 1.25 ms ASOI. b) 2.50 ms ASOI.

Figure 6 shows the imaging processing sequence for two selected timebins. As suggested in the post-processing section, a binary liquid image (figure 6 (i)) is used as a mask to ignore liquid fractions in the vapour image (figure 6 (ii)). Finally, the corrected image (figure 6 (iii)) is calibrated for fluorescence concentration measurements to obtain figure 6 (iv).

For the first timebin, 1.25ms ASOI, the injector is still fully open during the main injection stage. Therefore, such large amounts of liquid fractions close to the nozzle due to primary atomisation are expected, as can be seen in figure 6ai. However, figure 6aiii shows vapour in areas around the sac volume and toward the edges of the spray. At this distance downstream the spray is already in the secondary atomisation stage and as the droplets get smaller they are expected to evaporate more rapidly, creating vapour rich zones. The maximum relative concentration of fuel recorded in figure 6aiv was 0.32, and can be found around the edges of the spray. The stoichiometric fuel-to-air ratio for this tracer mixture is 0.067. So, although the experiments were conducted in a nitrogen atmosphere, the molecular weight of nitrogen can be approximated to the molecular weight of air, for which the measured concentration can be compared to the stoichiometric fuel-to-air ratio for indicative reasons. Allowing that comparison, a concentration fuel to nitrogen

concentration of 0.3 would correspond to an equivalence ratio around 4.5, which describes a very rich mixture. Figure 6bi confirms the presence of liquid droplets in the spray at 2.5ms ASOI. After correcting for liquid fluorescence, figure 6biii shows that most of the spray has already evaporated. The maximum concentration recorded in figure 6biv is about 0.23 in the front edge of the spray which gives an indicative equivalence ratio of 3.4.

Preliminary results presented in figure 6 show that the unambiguous separation of liquid signals provide valuable information about the location of liquid droplets in the vapour images, which allows correction of the vapour images for quantitative results. However, significant errors are introduced to the corrected images due to the comparatively large size of the pixels being disregarded by the binary liquid images. For instance, the final pixel size of these experiments is around 100 x 100 microns, so ignoring 1 pixel generates a hole of 100 x 100 microns in the corrected image, however, this pixel could be the result of the fluorescence of a single 40micron droplet, for which a much larger area than required is ignored. Therefore, a camera with extremely large pixel resolution needs to be used in order to apply this criterion for separation of vapour fluorescence, or a smaller area of interrogation should be considered.

## CONCLUSION

This paper discussed the applicability of a laser induced exciplex technique for quantitative measurements of G-DI sprays. Spectrally separated signals for liquid and vapour fractions were obtained with careful selection of filters. Preliminary results recorded at ambient conditions pertinent to in-cylinder conditions showed that the technique was successful describing qualitative liquid and vapour fractions. The main conclusions in this study are:

- An unambiguous separation of liquid signal was obtained with careful filter selection, and utilisation of a suitable tracer pair.
- The technique for correcting for the contribution of liquid fluorescence in the vapour images proved successful.
- A calibration curve was constructed for the test conditions showing a linear trend.
- Recommendations have been proposed for the errors introduced in the phase separation process due to the large size of the pixel in the experimental setup.
- Quantitative images of vapour concentration were obtained at ambient conditions relevant to in-cylinder conditions.
- This technique gives novel insight to the applicability of LIEF for quantitative measurements of vapour concentration / air-to-fuel ratios in the vicinity of spark plug for G-DI applications.

Future work will emphasize in studying the crosstalk of liquid and vapour signals at different pressures and temperatures. Also, the possibility of substituting benzene as one of the tracers will be explored due to its notorious hazardous association.

## ACKNOWLEDGMENT

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