Optical Fluorescence Auto-Projection Tomography: A Novel Modality

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Abstract *– We report preliminary research into a novel tomography system for application to gas-phase subjects. The technique is based on fluorescence, using collimated sources and detectors. It does not require a complex inversion step to reconstruct the image of the subject. Suitable gaseous fuels, dopants, and sources are assessed for potential use in a fuel imaging system within an internal combustion engine.*

Keywords: Fluorescence, Tomography, Fuel, Visualisation.

1 INTRODUCTION

A wide range of tomographic modalities have been developed over several decades, encompassing the classes of hard-field, soft-field and emission techniques[1]. All of these different classes have their various strengths and weaknesses in a range of applications. In particular, they all require an inversion calculation to reconstruct the distribution of the parameter of interest[2].

One particularly interesting option, which fits none of the above classes, is fluorescence tomography: the stimulation process has a “hard-field” nature in that the only material affecting the traversal of the stimulant beam through the subject is that which is in the geometrical path, whilst the isotropically emitted fluorescence clearly has an “emission” nature[3]. The early development of such a system is reported here.

As discussed elsewhere[4], spatial variation of air-fuel ratio in an internal combustion engine is of great commercial and environmental interest. Optical tomographic determination of this parameter has several potential advantages over other techniques, such as Planar Laser-Induced Fluorescence (PLIF)[5],[6]. Hence, the fuel-air mixture is the target subject for the present system development, and its specific requirements are discussed in section 2.

2 CONCEPT

2.1 General

In the case of X-Ray fluorescence, Cesareo and Mascarenhas[7] have pointed out that collimation of both the stimulation beam and the detector acceptance results in an unambiguous determination of the spatial region (hereinafter termed “space-point”) from which the signal was emitted. In their case, where they examined a relatively dense host, significant attenuation of both the stimulation beam and the fluorescence signals required an additional complexity in image reconstruction. The need to account for the subject’s attenuation can be removed by the selection of a subject with lower absorption, for example gaseous subjects diluted in air. The result is a simple geometrical reconstruction of the true space-points in the subject. We call this property “Auto-Projection”, since it is pre-determined by the geometry of the system and image reconstruction requires no inversion step.

Hence, the term “Auto-Projection Tomography” (APT) refers to the use of collimated sources and detectors to determine unambiguously a parameter of interest at a number of geometrically defined space-points within a larger subject of limited absorption. In the optical domain this can be achieved by exploiting the fluorescence of the subject to measure its concentration. The spatial intersection of a particular source beam and the acceptance of a particular detector define a unique space-point from which any signal contribution must have originated if no other source was active at the measurement instant. A system with N independently controllable sources and M detectors will provide NM space-points.

An Optical Fluorescence Auto-Projection Tomography (OFAPT) system with two sources and two detectors requires two measurement periods to resolve four space-points, see Figure 1. During the first period, source 1 is active while both detectors are sampled; in the second period source 2 is active while both detectors are sampled.

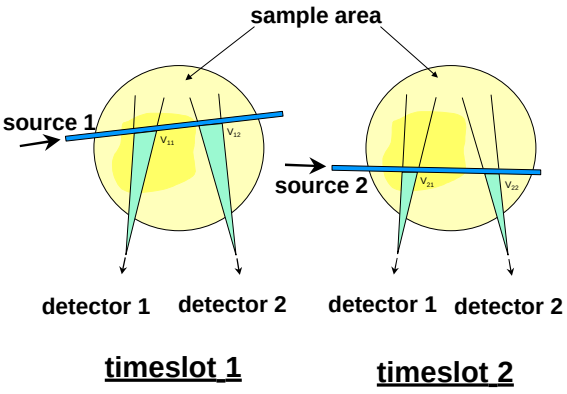

Figure 1: 2 x 2 Auto-Projection System.

Even moderate values of N and M can result in large numbers of space-points. Provided there are sufficient distributed space-points, then interpolation between the space-points will allow the image reconstruction of the entire subject.

The parameter reconstruction step[1] of more conventional tomographic modalities is more complex since each detector is sensitive to path-integrated contributions across the subject for hard-field modalities, or contributions from the whole of the subject (in principle) for soft-field modalities.

The critical requirements for an OFAPT system are that the fluorescence properties of the subject are well known, and that the sources have to be operated sequentially during the acquisition of data for a single image. The fundamental issue in developing this technique is the trade-off between the need for spatial and temporal resolution on the one hand, and the requirement to have a suitably strong and quantifiable fluorescence signal on the other. This is developed further in section 5.

2.2 Automotive R&D

The development of an OFAPT system has been focused on a fuel imaging system for internal combustion engines. PLIF is a widely used technique to visualise fuel within an engine: A laser is formed into a sheet, passed through the engine, and fluorescence is observed in an orthogonal direction, typically with a CCD camera[6],[8].

In the Stokes mechanism[9], incident photons are absorbed by molecules, resulting in emission of photons of a longer wavelength (red shifted) than the absorbed photons. This fluorescent emission is used to measure the subject's concentration via the relationship [10]:

$$I_F = 2.303 \times I_O . K . f_F . e . c . x \qquad (1)$$

$$\text{for } e.c.x < 0.05,$$

where:
I_O and I_F, are intensities of the incident and emitted light respectively, in units of mW mm^{-2};
The fluorescence properties of the subject are given by the Extinction Coefficient, ε (moles^{-1} litres cm^{-1}) and Quantum Efficiency, Φ_F;
c is the concentration of the subject (moles/litres) and x is the path length through the voxel (cm);
K is a geometrical and optical factor, which represents the collection efficiency for the emitted photons.

This mechanism has been exploited[11], [12] to measure species such as OH, O_2, and NO in flames, by the laser excitation of particular electronic transitions, and by wavelength-resolved detection.

In engine studies the subject is often the fuel or a dopant contained in the fuel[6]. The large number of pixels of CCD cameras and the short, high power laser pulse ensure that the spatial and temporal resolutions of the images captured are adequate. However the orthogonal optical access required is intrusive in the sense that severe modifications to the engine are necessary: large glass inserts are placed between the cylinder head and the engine block allowing the laser sheet to pass through the top of the combustion chamber; an elongated piston with a window in the centre is used with a mirror to provide the CCD with a "through-piston" view of the laser sheet. The result is a complex opto-mechanical solution which is difficult to implement even on research engines. The image framing rate of PLIF systems is severely restricted by the low repetition rates (0.1-100Hz)[5] of the lasers. Nevertheless PLIF systems do resolve real space-points.

The detectors in an OFAPT system are distributed around the cylinder in the same plane as the stimulation, therefore a reduction in the degree of optical access can be realised. Using optical fibres[13] to deliver the light to and from the combustion chamber can decrease intrusion further. A series of sequential light sources at a suitable wavelength will allow an OFAPT system to operate at an increased framing rate. The ideal is to capture consecutive frames each with a temporal resolution of 1° crank angle (CA) rotation for an engine running at say, 1000rpm. In such a case the time available for the operation of each of the N sources and the measurement of each space-point is, (167/N) μs.

3 FLUORESCENCE SPECTRA

3.1 General

The fluorescence of several chemical systems has been studied, ranging from 3-pentanone diluted in Iso-octane, to a range of commercial gasolines. In the selection[14] of a fluorescing subject the following properties must be considered.

- $\varepsilon(\lambda)$, absorption of the incident beam.
- $\phi_F(\lambda)$, conversion efficiency of absorbed photons to fluorescence photons.
- Magnitude of the Stokes red shift.
- Emission dependence upon temperature and pressure.
- Fluorescence quenching by oxygen.

The extinction coefficient and quantum efficiency are both functions of excitation wavelength, and their product effectively determines the realistic ranges within which the stimulation and detection systems must operate. The red shift exhibited by a subject must be sufficient to allow filtration of the emitted and/or stimulation light, thereby minimising any elastically scattered stimulation light seen by the detection system. Pressure, temperature, oxygen quenching and self re-absorption may significantly influence the detected fluorescence intensity. The majority of information which may be obtained from an engine study will be at the maximum compression of the cylinder (≈1MPa for a spark ignition engine) and high temperature (≈400K).

3.2 Dopants

If the fuel used is composed of a non-fluorescing base and a fluorescing dopant then the dopant must be readily soluble in the base and have a similar boiling point. Such a fuel will allow the desired fuel absorption to be achieved by controlling the dopant concentration.

3-Pentanone ($C_5H_{10}O$) is often used in PLIF studies to dope Iso-octane (C_8H_{18}). Research grade or HPLC Iso-octane does not fluoresce [6],[8]. Hansen[15] has determined the extinction coefficient, quantum efficiency and emission of 3-Pentanone. The variation of these properties with temperature, pressure, and oxygen quenching, have been studied[6],[8] and found to be favourable for combustion studies in which Iso-octane is the base fuel. (This is a commonly used fuel in engine and combustion research and development.) It is worth noting that the peak absorption of 3-Pentanone occurs at 285nm and peak emission at 430nm. The quantum efficiency is effectively independent of wavelength, therefore the excitation spectra will have the same shape as the absorption spectra. This is provided for comparison with the excitation spectra of gasoline in Figure 2. Excitation spectra represent the ability of a species to absorb stimulation photons and emit fluoresced photons, as a function of stimulation wavelength. They may be measured using a fluorimeter with a fixed emission wavelength and scanning a range of stimulation wavelengths.

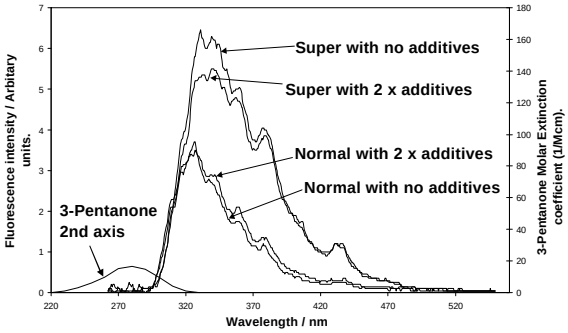

Figure 2: Excitation spectra of Gasoline compared with the absorption of 3-Pentanone.

3.3 Gasolines

The excitation spectra for a number of gasolines were determined, see Figure 2. The gasolines were diluted in 10 parts of HPLC grade Iso-octane. A typical gasoline contains approximately 35% aromatics. These components can be studied and the fluorescence properties of each assessed[16].

Aromatics	Boiling Point °C	Quantum Efficiency
Benzene	80	0.07
Toluene	111	0.17
Ethyl benzene	136	0.18
Meta-xylene	138	0.17
Para-xylene	138	0.40
Ortho-xylene	144	0.19
1,3,5-trimethylbenzene	170	0.12
1,2,4-trimethylbenzene	148	0.41
3-Pentanone	102	0.0025

Table 1: Properties of typical aromatic species found in gasoline, along with 3-Pentanone data for comparison.

The data in Table 1 show that gasoline contains species with quantum efficiency two orders of magnitude larger than that of 3-Pentanone. The higher fluorescence signal from 'Super' grade gasoline, compared with 'Normal' in Figure 2, is due to the higher aromatic content. The red shift observed in these species is significantly smaller than for 3-Pentanone and the use of interference filters is likely to be

required to exploit them. The fluorescence properties of a full gasoline mixture in a combustion environment have been found to be less favourable than for 3-Pentanone[14], i.e. in terms of oxygen quenching, re-absorption, pressure and temperature effects. A severe decrease in emission was experienced at the elevated pressures in a running engine.

4 TECHNOLOGY OPTIONS

4.1 Sources

The nature of the optical sources for stimulation of fluorescence is the most critical aspect of OFAPT system design. The source requirements for an OFAPT system are:

- Multiple sources
- Sequential operation
- Wavelength in a suitable range
- Adequate power
- Fibre coupling for engine application

The requirement for sequential operation can be achieved by a number of discrete sources centrally controlled, one source scanned over multiple light channels, or one source divided equally into a number of light channels with intensity modulation of each channel. An OFAPT system with 10 sources must be able to operate with a maximum pulse duration of 17μs (see section 2.2).

The sequential operation and UV wavelength required for the stimulation of 3-Pentanone (285±25nm) present considerable difficulty in identifying a suitable OFAPT source. In the gasoline case, fluorescent species are normally present (although generally less well understood than 3-Pentanone), which yield strong signals upon stimulation over a wider range of wavelengths. Sources in the range of 300-500nm may be considered for the stimulation of gasoline, as seen in Figure 2.

For OFAPT, the critical parameter for any source is the total energy delivered, $E_{P,}$ for each fluorescence sampling period. We use this parameter below, for a sampling period of 17μs.

Discharge Lamps

Deuterium Lamps[17] provide broadband emission from 200nm to 400nm. A complex drive circuit is required for sequential operation of multiple lamps. Optical filtering would be necessary to remove the visible component of the lamps' output.

Higher power Xenon discharge sources could be implemented as a single source scanned over a number of fibres or coupled to multiple fibres. Scanning could be achieved using a rotating mirror. Alternatively a multi-faceted opto-mechanical chopper rotating at approximately 90 Hz could modulate multiple fibres coupled to one lamp. The scanning and chopping solutions would both result in a complex opto-mechanical construction along with a significant insertion loss. Neglecting insertion losses, we calculate these sources yield E_P=340nJ.

Semiconductor Sources

Semiconductor sources offer the most convenient method of providing the sequential source operation required in an OFAPT system. Unfortunately there are no semiconductor devices available at present capable of providing UV radiation for the stimulation of 3-Pentanone.

Blue Laser Diodes[18] have been reported by Nichia, yielding 50mW at 410nm. These could be considered for use in a final system but the schedule for commercial availability of the diodes is not clear. These devices promise E_P=850nJ.

LEDs are low cost, low power discrete sources, available in the blue region with nominal wavelengths of 430 and 470nm, half width of 40nm, and approximately 2mW power output. Although difficult to couple to fibre efficiently, they are convenient to drive sequentially. Pulsed operation with a low duty cycle can allow an increase in the available peak power by a factor of 10 or so. We calculate E_P=340nJ.

Pulsed Lasers

The Nd:YAG and Excimer lasers[5] used in conventional PLIF studies produce very intense (10-100mJ) short (10ns) pulsed outputs at a wavelength suitable for 3-Pentanone excitation. However they operate at a typical repetition rate of 1 Hz with a maximum of 100Hz, which is two to three orders of magnitude slower than required for the 1°CA frame rate target. Typically, E_P=10mJ.

A Ce:LiCAF[19] laser pumped with a copper vapour laser can yield up to 450mW average power output at a wavelength of 288.5nm with a repetition rate of 7kHz and a 8ns pulse length, yielding E_P=64μJ. Whilst the requirement of a copper vapour laser and custom built non-linear optical apparatus is significant, this option has promise if in-fibre multiplexing can be achieved.

Frequency-quadrupled Nd:YAG lasers are capable of producing high average powers with repetition rates above 5kHz[20]. Operating at 1kHz an average power of 2.8W was obtained at 266nm i.e. E_P=2.8mJ. As this laser is not presently available as a commercial product, a custom non-linear optical setup must be

constructed. Nevertheless, the E_P value for this source is very attractive.

Continuous Wave Lasers

Gas lasers at suitable wavelengths are 325, 442nm (HeCd) and 457, 488nm (Argon Ion). These lasers can deliver approximate powers of 1mW to 1W, yielding E_P=17nJ to 17μJ . Electro-Optic or Acousto-Optic deflectors may be used to scan the laser over a number of fibres. Such deflectors are commercially available along with the necessary high-frequency drive electronics. Electro-Optic deflectors capable of producing deflections in the region of 1mrad/kV have been demonstrated[21] and should be suitable for use down to 300nm. Acousto-Optic deflectors are available for use down to 360nm.

Blue frequency-doubled lasers producing 15mW at 430nm (E_P=255nJ) are commercially available and have very similar characteristics to the blue laser diode presently under development by Nichia. This laser may be modulated directly.

4.2 Optics

Launch

The characteristics of the launch optics will depend greatly on the source and modulation technique selected. The ideal performance of the launch optics would be to provide a collimated beam with a known path across the chamber. Provided the field of view of the detection system is not exceeded, the total fluorescence signal yield is independent of the source cross section area. Therefore a badly collimated beam can be tolerated, within the limits of the system spatial resolution and the detector field of view.

Receive

The collection optics of the whole measurement system are required to gather fluorescence from voxels, over the entire subject. The collection optics for each detector must therefore have a limited field of view. The degree of restriction will define the spatial resolution and the magnitude of any measurements made. An additional problem to consider is the dependence of the signal magnitude upon the distance from the voxel to the detection lens. This can be expressed within the factor K, in terms of the proportion of the isotropically emitted fluorescence which can be collected. If a conical collection scheme is employed, the voxel-to-lens distance dependence is reduced. However the voxel size and hence spatial resolution of the system then become functions of the voxel-to-lens distance.

Detectors

The minimum resolvable power levels of the detectors will directly influence the total stimulation power required to perform the fuel concentration measurements with the desired resolution and signal to noise ratio. Each detector and associated amplifier must have a sufficient bandwidth to allow sampling during the active period of each source, with appropriate noise suppression.

5 THEORETICAL CALCULATIONS

The factors influencing the intensity of observed fluorescence have been incorporated into a calculation scheme which, for a given optical configuration, yields the ratio of fluorescence intensity to stimulation intensity (I_F/I_O) see equation (1). For the purposes of illustration, the total path absorption of a stoichiometric homogenous mixture was defined as 5%, which is a reasonable value for various cases. The path was divided into r voxels (r=8, 16, or 32) and the absorption of each voxel determined for the above conditions using the following relationship between path and voxel transmissions.

$$T_{voxel} = (T_{path})^{1/r} \quad \textbf{(2)}$$

Figure 3 shows I_F/I_O for a measurement of a voxel at the centre of an 80mm diameter combustion chamber, using a 1.5mm diameter collection lens and Iso-octane doped with 3-Pentanone (Φ_F=0.0025). The concentration of 3-Pentanone is fixed by the voxel absorption obtained from equation (2) above.

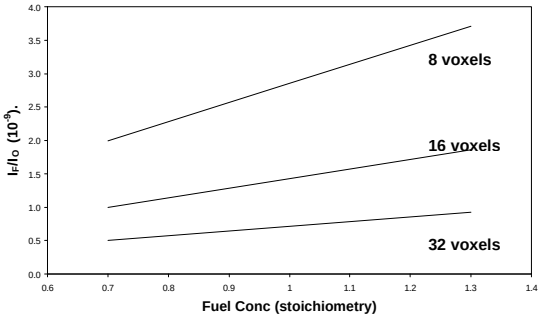

Figure 3: I_F/I_O per voxel vs. Fuel Conc., with the path divided into various numbers of voxels (total path absorption 5%).

With knowledge of I_F/I_O, and the minimum resolvable power of the detection system, the stimulation power required to perform the fuel

concentration measurements with the desired accuracy and resolution can then be determined. To view the above in the context of the present application it should be noted that, a stoichiometric mixture at 1MPa and 400K will give a total path absorption of 10% (λ=285nm) with a 19% dopant to fuel ratio; this is typical [6]. Note also that the fluorescence yield of any other species in the above calculation will scale simply in the ratio of the quantum efficiencies, neglecting any quenching effects.

The following assumptions have been made:

- No light is scattered during traversal to or from the fluorescing voxel.
- The voxel is stimulated via one face and fluorescence detected via an orthogonal face.
- The Fluorescence source is a point in the centre of the voxel.
- Fluorescence saturation is not reached.
- The Extinction coefficient for 3-Pentanone is dominant.

The I_F/I_O values in Figure 3 are of the order of 10^{-9}, which indicates that a very intense source (≈1W) would be necessary in order to apply this technique for 3-Pentanone/Iso-octane in the present case.

6 EXPERIMENTAL WORK

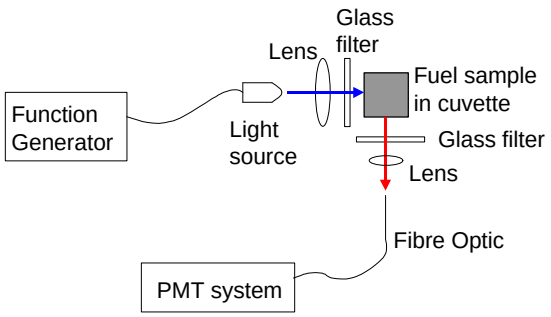

Figure 4: Liquid Phase Fluorescence system

As a preliminary step to gas-phase experiments, a simple system was established to measure the fluorescence of a liquid phase sample contained within a cuvette, Figure 4. The system was operated with either a bright blue LED or a Deuterium lamp. Effects of scattered light were minimised by inserting appropriate glass filters, and using perpendicular source and detection light paths. Fluoresced light could be delivered to the photomultiplier tube by either a 200 or 500μm diameter fibre optic.

Gasoline

The signal obtained from a gasoline sample diluted in Iso-octane stimulated by a blue LED, was directly confirmed to be fluorescence by performing a spectral analysis of both the source and the emitted light using a computer-controlled scanning monochromator. The spectra in Figure 5 show the red shifted fluorescence signal compared with the LED spectrum.

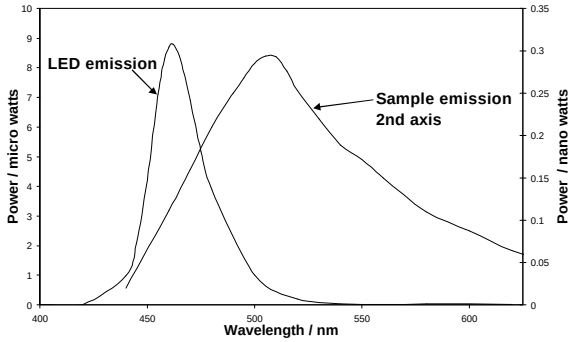

Figure 5: Spectral analysis of blue LED and gasoline fluorescence

As expected, increased emissions were observed at higher gasoline concentrations. The LED peak emission wavelength of 460nm corresponds to a weak absorption region on the gasoline excitation spectrum, as shown in Figure 2. A shorter wavelength source would yield a stronger fluorescence signal per unit of stimulation power. Blue LEDs are capable of producing fluorescence from gasoline, but are unlikely to provide sufficient fibre-coupled power for an OFAPT system where the gaseous sample concentration is 3 orders of magnitude lower than for a liquid, and the measurement voxel is distant from the collection lens. Alternatively, more intense blue sources, as discussed in section 4.1, may provide a desirable solution.

3-Pentanone

A deuterium lamp provided UV radiation at 285±25nm to stimulate 3-Pentanone. The lamp output was filtered using a glass UG5 filter to remove wavelengths longer than 410nm. A suitable dopant concentration was determined by performing a number of transmission tests using a deuterium lamp, a Bentham monochromator and a Newport power meter. Weak fluorescence was observed with a photomultiplier tube. Brighter lamps with smaller apertures are available and are 4 times brighter. Lamp intensities can be further increased by pulsing the lamps at a low duty cycle, allowing an additional increase up to 7 times in lamp intensity. Although a small-aperture, pulsed, fibre-coupled deuterium lamp will give a reasonable fluorescence signal with 3-

Pentanone, the intensity is too low for implementation into a real system where the measurement voxel may be located in the centre of the combustion chamber.

Light source	Subject	Fibre diameter (μm)	Signal captured (photon/μs)
LED	Gasoline	500	1027
LED	Gasoline	200	86
D2	3-Pent	500	6
D2	Gasoline	500	216

Table 2: Liquid fluorescence results

All the liquid phase experiments were conducted with an identical optical arrangement and detection system, Figure 4. The results from the liquid phase experiments (Table 2) show that the intensity of the 3-Pentanone fluorescence was much weaker than that of gasoline, as expected. The geometrical coupling in the liquid phase experimental system is significantly higher than would be expected in a real system where the voxel to detector distance may be the radius of a combustion chamber (≈40mm). The voxel to detector distance in the liquid phase experiment was approximately 8mm, implying a signal reduction factor of 25 if the distance is increased to 40mm.

7 FUTURE WORK

The behaviour of a single channel system is fundamental to the entire OFAPT system. Vapour phase experimental work presently underway will provide information concerning:

- Signal yield (per unit power stimulation light) for a gas phase subject under similar conditions to that of a spark ignition combustion engine.
- Dependence of the above upon temperature and pressure.
- Oxygen quenching effects, which may be severe for the gasoline case[14].

An estimate of the minimum power levels required at a given wavelength for an OFAPT system, with the desired signal to noise ratio, can be made more confidently, based on this type of experimental data. The sources still under consideration are broadband UV discharge (Xe) lamps and blue lasers. Once a single channel system has been completed this may form the basis of an entire system to be implemented on a research engine, see Figure 6.

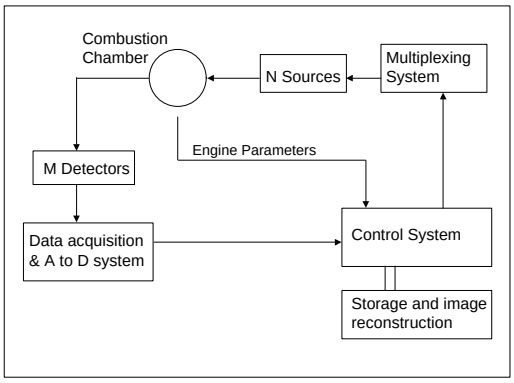

Figure 6: Schematic of an entire OFAPT system, in an engine application.

8 CONCLUSIONS

An OFAPT system has the potential to resolve real space-points and perform image reconstruction using a simple algorithm. The suitability of the light source for the fluorescing species dictates the characteristics of a fluorescence based measurement system. At present, ideal light sources do not exist for OFAPT combustion chamber fuel imaging.

Deuterium lamps are not powerful enough for implementation into a real OFAPT system. Existing blue laser sources are not optimally suited to the absorption spectrum of gasoline. Blue semiconductor lasers appear promising in the medium term. In the longer term, pulsed lasers may provide a possible solution.

There are many advantages in the use of a 3-Pentanone tracer. However, its low quantum efficiency prevents its use with low intensity sources of light. The fluorescence of gasoline is less quantifiable than that of 3-Pentanone. Gasoline fluorescence will not only vary from one batch to another but is likely to suffer from quenching and re-absorption especially at high pressure. OFAPT does appear feasible with presently available light sources for applications with less demanding imaging rate requirements, i.e. where sampling periods in the region of 10ms would be suitable.

The design of a system must ensure that I_F/I_O is optimised; the use of a conical detection path will assist this but will result in a non-homogeneous spatial resolution which must be reflected in the reconstruction algorithm, assigning greater confidence to the measurements with better spatial resolution.

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