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DUAL EMISSION LASER INDUCED FLUORESCENCE TECHNIQUE (DELIF) FOR OIL FILM THICKNESS AND TEMPERATURE MEASUREMENT

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ABSTRACT

This paper presents the development and implementation of a Dual Emission Laser Induced Fluorescence (DELIF) technique for the measurement of film thickness and temperature of tribological flows. The technique is based on a ratiometric principle that allows normalization of the fluorescence emission of one dye against the fluorescence emission of a second dye, eliminating undesirable effects of illumination intensity fluctuations in both space and time. Although oil film thickness and temperature measurements are based on the same two-dye ratiometric principle, the required spectral dye characteristics and optical conditions differ significantly. The effects of emission reabsorption and optical thickness are discussed for each technique. Finally, calibrations of the system for both techniques are presented along with their use in measuring the oil film thickness and two-dimensional temperature profile on the lubricating film of a rotating shaft seal.

INTRODUCTION

Laser Induced Fluorescence (LIF) is based on the use of a light source to excite a fluorescence substance (fluorophore or fluorescent dye) that subsequently emits light. The fluorescence substance is used as a tracer to determine characteristics of interest. LIF has gained popularity as a general purpose visualization tool for numerous 1-D, 2-D, and 3-D applications. It, however, has seen limited use as a quantitative tool. The reason for this stems primarily from the difficulty in separating variations in excitation illumination and vignetting effects from tracer emission. Presented herein is a two-dye ratiometric technique that allows measurement of temperature and film thickness while minimizing variations in excitation illumination and non-uniformities in optical imaging.

Fluorescence is the result of a three-stage process that occurs in fluorophores or fluorescent dyes (Haugland, R. P., 1999). The three processes are (Fig. 1):

1: Excitation

A photon of energy hv_{EX} is supplied by an external source such as an incandescent lamp or a laser and absorbed by the fluorophore, creating an excited electronic singlet state (S_1').

2: Excited-State Lifetime

The excited state exists for a finite time (typically 1–10 x 10^{-9} seconds). During this time, the fluorophore undergoes conformational changes and is also subject to a multitude of possible interactions with its molecular environment. These processes have two important consequences. First, the energy of S_1' is partially dissipated, yielding a relaxed singlet excited state (S_1) from which fluorescence emission originates. Second, not all the molecules initially excited by absorption (Stage 1) return to the ground state (S_0) by fluorescence emission. Other processes such as collisional quenching, fluorescence energy transfer and intersystem crossing may also depopulate S_1. The fluorescence quantum yield, which is the ratio of the number of fluorescence photons emitted (Stage 3) to the number of photons absorbed (Stage 1), is a measure of the relative extent to which these processes occur.

3: Fluorescence Emission

A photon of energy hv_{EM} is emitted, returning the fluorophore to its ground state S_0. Due to energy dissipation during the excited-state lifetime, the energy of this photon is lower, and therefore of longer wavelength, than the excitation photon hv_{EX}. The difference in energy or wavelength represented by (hv_{EX}–hv_{EM}) is called the Stokes shift. The Stokes shift is fundamental to the sensitivity of fluorescence techniques because it allows emission photons to be detected against a low background, isolated from excitation photons.

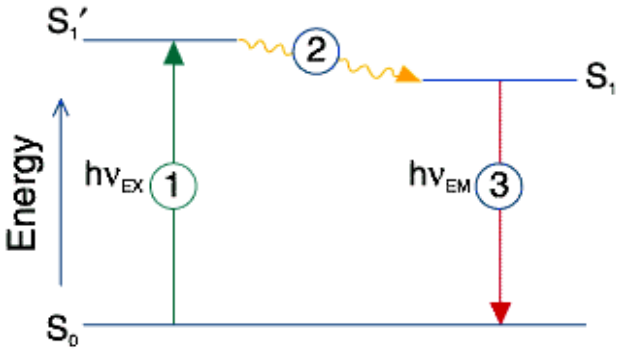


Figure 1: Fluorescence principle (Haugland, R. P., 1999)

From this description it is apparent that LIF can be used to measure any scalar that affects the fluorescence of the dye. Fluorescence is a function of the dye characteristics, the dye concentration, the exciting light intensity, and the scalar being measured. Once a particular dye and concentration are selected, the fluorescence dependence on these factors is constant. The problem lies in the irregularity of the illumination light intensity when a laser is used. Most laser beams are not uniform. They fluctuate in intensity in space and time. Pulsed Nd:YAG lasers are particularly prone to exhibit this behavior. Use of pulsed Nd:YAG lasers is desirable though, because of their short pulse duration (consequently short fluorescence emission), which allows for nearly instantaneous measurements of the desired scalar.

In order to correlate two-dimensional fluorescence intensity to the scalar of interest, spatial variations in illumination intensity must be determined. This can be accomplished by using a ratiometric technique where the fluorescence intensity containing the desired scalar information is divided by the laser intensity eliminating the fluorescence dependency on excitation intensity. One way to achieve this is by using two fluorescent dyes. This technique is known as Dual Emission Laser Induced Fluorescence (DELIF) (Coppeta, J., Rogers, C., 1998, Coppeta, J., et. al., 1997, Sakakibara, J., Adrian, R. J., 1999).

NOMENCLATURE

A	area of one pixel
C	dye molar concentration, effective two-dye molar concentration
C_1	dye 1 molar concentration
C_2	dye 2 molar concentration
dI_f	differential fluorescence intensity
$dI_{f,1}$	dye 1 differential fluorescence intensity, without reabsorption
$dI_{f,1}'$	dye 1 differential fluorescence intensity, with reabsorption
dx	differential length in x-direction
F	fluorescence power
I_e	exciting light intensity
I_f	total fluorescence intensity
$I_{f,1}$	dye 1 total fluorescence intensity, without reabsorption
$I_{f,1}'$	dye 1 total fluorescence intensity, with reabsorption
$I_{f,2}$	dye 2 total fluorescence intensity
I_o	exciting light intensity at x=0
t	film thickness
T	temperature
x	coordinate perpendicular to plane of observation
y	coordinate parallel to plane of observation
ΔV	volume element
$\varepsilon(\lambda)$	molar absorption (extinction) coefficient at a given wavelength (absorption spectrum); effective two-dye molar absorption (extinction) coefficient
$\varepsilon_1(\lambda)$	dye 1 molar absorption (extinction) coefficient at a given wavelength (absorption spectrum)
$\varepsilon_2(\lambda)$	dye 2 molar absorption (extinction) coefficient at a given wavelength (absorption spectrum)
Φ	quantum efficiency
Φ_1	dye 1 quantum efficiency
Φ_2	dye 2 quantum efficiency
$\eta_1(\lambda)$	dye 1 relative emission at a given wavelength (emission spectrum)
$\eta_2(\lambda)$	dye 2 relative emission at a given wavelength (emission spectrum)
λ_{laser}	laser wavelength
$\lambda_{filter1}$	narrow band filter 1 wavelength
$\lambda_{filter2}$	narrow band filter 2 wavelength
τ	time

LIF BASICS

Optically Thin versus Optically Thick

Consider a rectangular differential volume of fluid mixed with a fluorescent dye with cross-sectional area A and length Δx irradiated by light (normal to the area A) with uniform intensity I_e (see figure 2). The total fluorescence, F, emitted by this differential volume is given by:

$$F = I_e \varepsilon(\lambda_{laser}) C \Phi \Delta V \qquad (1)$$

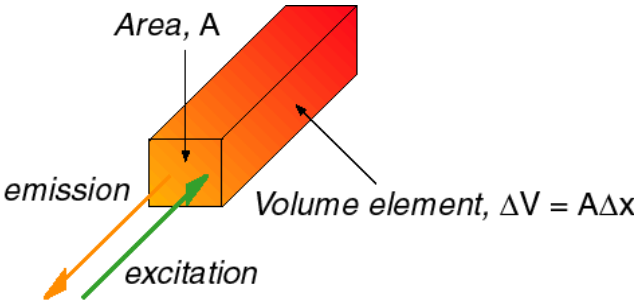


Figure 2: Fluorescence of fluid element

From equation (1), it is evident that fluorescence intensity is dependent on:

(1) the amount of exciting light available to produce molecular transitions to higher, excited levels,

(2) molar absorpsivity, which determines how much of the incident light per molecule produces actual molecular transitions,

(3) dye concentration, which is a measure of the number of molecules present,

(4) quantum efficiency, which is the ratio of the energy emitted by the energy absorbed, and is a measure of how much of the energy stored in the higher electronic states is emitted as fluorescence light, when the molecules return to their ground state, and,

(5) the volume of the element, which is the control volume over which excitation and fluorescence takes place.

Dividing equation (1) by the area *A*, the fluorescence intensity normal to the area *A* is obtained,

$$I_f = I_e \varepsilon(\lambda_{laser}) C \Phi \Delta x \qquad (2)$$

If the area *A* is assumed to be the projected area of a single pixel, it is apparent that pixel intensity is proportional to the excitation intensity, dye characteristics, concentration, and thickness of the fluid element. For very thin film thickness, this representation is accurate. If the excitation intensity is known, dye characteristics, and concentration are constants, the fluid film thickness can be directly inferred from the fluorescence. A more accurate representation of the fluorescence phenomena can be obtained by from Lambert's Law of Absorption (Poll, G., et. al., 1992), which takes into account the absorption of the exciting light by the finite fluid through which it travels;

$$I_e(x) = I_o \exp\left[-\varepsilon(\lambda_{laser}) C x\right] \qquad (3)$$

Consider the differential element shown in figure 3 within a region of finite film thickness. The fluorescence intensity collected by the CCD from this fluid element is

$$dI_f = I_e \varepsilon(\lambda_{laser}) C \Phi dx \qquad (4)$$

.

Thus, from equation (4):

$$dI_f = I_o \exp\left[-\varepsilon(\lambda_{laser}) C x\right] \varepsilon(\lambda_{laser}) C \Phi dx \qquad (5)$$

For a given fluid thickness, *t*, the total intensity collected by the CCD is

$$I_f(t) = \int_0^t dI_f = \int_0^t I_o \exp\left[-\varepsilon(\lambda_{laser}) C x\right] \varepsilon(\lambda_{laser}) C \Phi dx \qquad (6)$$

such that

$$I_f(t) = I_o \Phi \left\{1 - \exp\left[-\varepsilon(\lambda_{laser}) C t\right]\right\} \qquad (7)$$

For small values of *t* (thin films), equation (7) can be approximated as:

$$I_f(t) \approx I_o \varepsilon(\lambda_{laser}) C \Phi t \qquad (8)$$

This is identical to equation (2) and is the basis for the concepts of optically thin and optically thick systems. The fluorescence dependence on film thickness is linear for optically thin systems, while it is exponential for optically thick systems. What is considered a thin or thick film thickness depends on the product $\varepsilon(\lambda_{laser})C$.

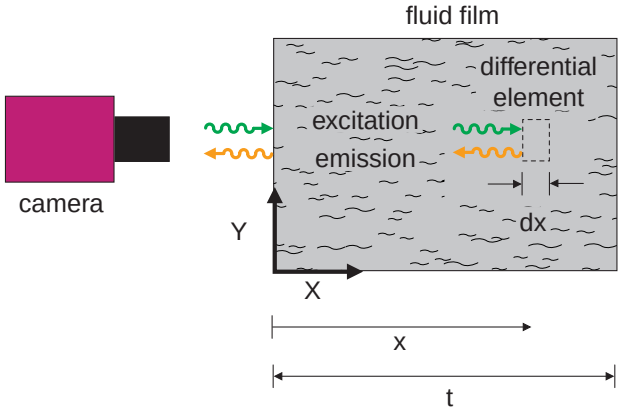


Figure 3: Fluorescence through a thick fluid film

Reabsorption

Emission reabsorption is often encountered in fluorescence techniques and is generally problematic. Fluorescent dyes have different absorption spectrums and emission spectrums (Fig. 4). When the emission spectrum of one dye overlaps the absorption spectrum of another or with its own absorption spectrum, reabsorption of the dye fluorescence occurs (Fig. 5). This has two effects: (1) it increases the fluorescence emission of the second dye as, in addition to the external light source excitation, it is being excited by the fluorescence of the first dye. More importantly, (2) the fluorescence emission of the first dye is reduced since it is being reabsorbed by the second dye. In LIF, the external illumination intensity is generally much greater than dye fluorescence. Consequently, the increase in fluorescence emission due to excitation by the fluorescence of

one dye by another can be neglected. This is not the case for the reduction in fluorescence of a dye due to reabsorption by a second dye since this reduction can be substantial in comparison with the total emission of the dye when there is no reabsorption. From the differential element of figure 3, it is apparent that the differential fluorescence emission produced by any single element must travel back through the medium before reaching the CCD.

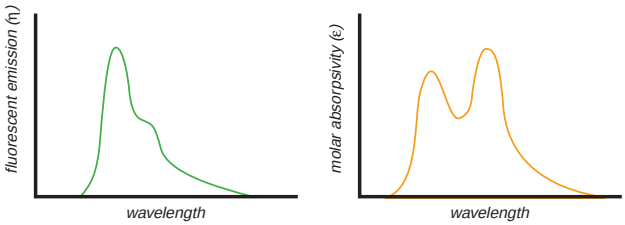


Figure 4: Emission and absorption spectrums

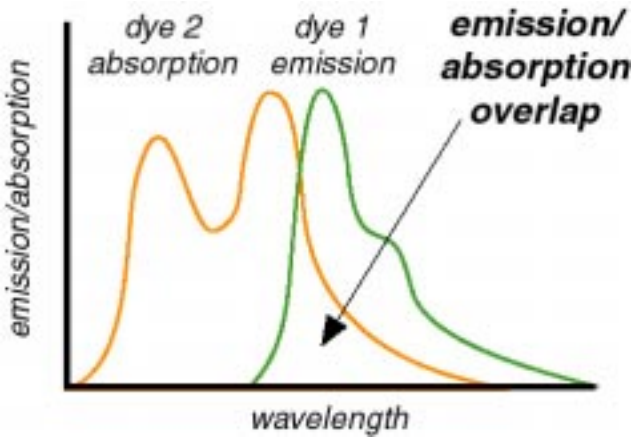


Figure 5: Reabsorption schematic

If there is reabsorption of the differential element fluorescence, Lambert's law must be applied to the differential fluorescence emission in order to compute the actual fluorescence collected by the CCD. Thus, assuming the situation represented in figure 5 occurs,

$$dI_{f,1}=I_o \exp\left[-\varepsilon(\lambda_{laser})Cx\right]\varepsilon_1(\lambda_{laser})C_1\Phi_1\eta_1(\lambda)\,dx\,d\lambda \quad (9)$$

$$dI_{f,1}'=dI_{f,1}\exp\left[-\varepsilon_2(\lambda)C_2 x\right] \quad (10)$$

$$dI_{f,1}'=I_o \exp\left[-\varepsilon(\lambda_{laser})Cx\right]\varepsilon_1(\lambda_{laser})C_1\Phi_1\eta_1(\lambda)$$

$$\times\exp\left[-\varepsilon_2(\lambda)C_2 x\right]dx\,d\lambda \quad (11)$$

Equation (5) has been modified in equation (9) to reflect the fact that the fluorescence emission occurs over a wide range of wavelengths that constitute the emission spectrum. In the same way equations (10) and (11) portray a reabsorption that occurs over a wide range of wavelengths. If the emission spectrum of dye 1 and the absorption spectrum of dye 2 are known, equation (11) can be integrated over varying film thickness and wavelengths in order to compute the total intensity collected by the CCD. If a very narrow interference filter is used to filter all wavelengths except for the one of interest, equation (11) can be simplified by removing the dependence of the differential intensity on the emission and absorption spectrums. Thus, the total intensity collected on the CCD can be calculated as:

$$I_{f,1}'(t\lambda_{filter1})=\int_0^t I_o \exp\left[-\varepsilon(\lambda_{laser})Cx\right]\varepsilon_1(\lambda_{laser})C_1\Phi_1$$

$$\times\eta_1(\lambda_{filter1})\exp\left[-\varepsilon_2(\lambda_{filter1})C_2 x\right]dx \quad (12)$$

$$I_{f,1}'(t\lambda_{filter1})=\frac{I_o\varepsilon_1(\lambda_{laser})C_1\Phi_1\eta_1(\lambda_{filter1})}{\varepsilon(\lambda_{laser})C+\varepsilon_2(\lambda_{filter1})C_2}$$

$$\times\left(1-\exp\left\{-\left[\varepsilon(\lambda_{laser})C+\varepsilon_2(\lambda_{filter1})C_2\right]t\right\}\right) \quad (13)$$

For reabsorption to play a significant role on the fluorescence, the system must be optically thick and $\varepsilon_2(\lambda_{filter1})C_2 >\approx O[\varepsilon(\lambda_{laser})C]$.

DELIF, THE RATIOMETRIC APPROACH

Principle

In the previous analysis, based on figure 3, and in equations (1) through (13), the non-uniformity of the exciting light intensity over the plane of observation and in time is not taken into consideration. In reality, illumination intensity is a function of both position and time;

$$I_o = I_o(y,\tau) \quad (14)$$

Therefore,

$$I_f = I_f(t,\lambda, y,\tau) \quad (15)$$

Consequently, film thickness cannot be inferred from fluorescence intensity unless illumination intensity at a particular location and time is known. The ratio of the illumination intensity and the fluorescence intensity, however, is independent of spatial and temporal variations in excitation light intensity.

$$\frac{I_f}{I_o} \equiv R = R(t,\lambda) \qquad (16)$$

Obtaining illumination intensity is not trivial. A two-dimensional instantaneous illumination map, however, can be inferred from the fluorescence of a second dye. This is the principle behind DELIF:

(1) the fluorescence of dye 1 in a two-dye system contains the desired information (film thickness, temperature, which will be discussed later), along with exciting light intensity information.

(2) the fluorescence of dye 2 also contains the exciting light intensity information but behaves differently than dye 1 to the scalar of interest.

(3) By rationing the fluorescence of dye 1 with the fluorescence of dye 2, the excitation light information is canceled out, giving a ratio that contains only the desired scalar information.

Oil Film Thickness

Oil film thickness measurements are achieved using an optically thick system that takes advantage of reabsorption. The film thickness information is contained in the reabsorption of the fluorescence of dye 1 by dye 2. The excitation light intensity information is contained both in the fluorescence of dye 1 and dye 2. If two narrow-band interference filters are used to capture the two distinctive fluorescence emissions, an emission intensity defined by

$$I_{f,1}'(t,\lambda_{filter1}, y, \tau) = \frac{I_o(y,\tau)\,\varepsilon_1(\lambda_{laser})C_1\Phi_1\eta_1(\lambda_{filter1})}{\varepsilon(\lambda_{laser})C+\varepsilon_2(\lambda_{filter1})C_2}$$

$$\times\left(1-\exp\left\{-\left[\varepsilon(\lambda_{laser})C+\varepsilon_2(\lambda_{filter1})C_2\right]t\right\}\right) \qquad (17)$$

$$I_{f,2}(t,\lambda_{filter2}, y, \tau) = \frac{I_o(y,\tau)\,\varepsilon_2(\lambda_{laser})C_2\Phi_2\eta_2(\lambda_{filter2})}{\varepsilon(\lambda_{laser})C}$$

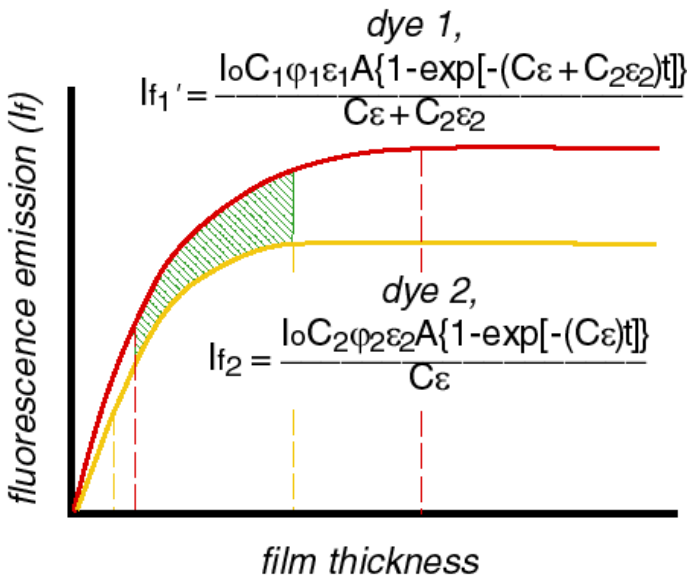
$$\times\left(1-\exp\left\{-\left[\varepsilon(\lambda_{laser})C\right]t\right\}\right) \qquad (18)$$

$$R(t,\lambda_{filter1},\lambda_{filter2}) = \frac{I_{f,1}'}{I_{f,2}} = \frac{\varepsilon_1(\lambda_{laser})C_1\Phi_1\eta_1(\lambda_{filter1})}{\varepsilon_2(\lambda_{laser})C_2\Phi_2\eta_2(\lambda_{filter2})}$$

$$\times\frac{\varepsilon(\lambda_{laser})C\left(1-\exp\left\{-\left[\varepsilon(\lambda_{laser})C+\varepsilon_2(\lambda_{filter1})C_2\right]t\right\}\right)}{\left[\varepsilon(\lambda_{laser})C+\varepsilon_2(\lambda_{filter1})C_2\right]\left(1-\exp\left\{-\left[\varepsilon(\lambda_{laser})C\right]t\right\}\right)} \qquad (19).$$

is obtained.

By taking the ratio of the emission of the two dyes, the excitation light intensity dependence is cancelled leaving a ratio that is only dependent on film thickness. As film thickness information is contained in the reabsorption of the fluorescence of dye 1 by dye 2, the system must be optically thick, in order for the reabsorption to be substantial and measurable (Fig. 6).



$$R(t) = \frac{I_{f1}}{I_{f2}} = \frac{C_1\varphi_1\varepsilon_1(C\varepsilon)\{1-\exp[-(C\varepsilon+C_2\varepsilon_2)t]\}}{C_2\varphi_2\varepsilon_2(C\varepsilon+C_2\varepsilon_2)\{1-\exp[-(C\varepsilon)t]\}}$$

Figure 6: Film thickness ratio

Temperature

It is possible to use LIF as a temperature indicator when there is a dependence of either the molar absorption (extinction) or quantum efficiency coefficients on temperature.

$$\varepsilon = \varepsilon(T) \qquad (20)$$

and/or

$$\Phi = \Phi(T) \qquad (21)$$

The problem of separating the temperature information from the exciting light intensity contained in the fluorescence still exists. This is further complicated by the film thickness information that is also imbedded in the fluorescence. The same two-dye fluorescence ratiometric approach used to separate the film thickness information from the exciting light intensity information can be used for temperature measurement. However, the optical conditions for proper temperature measurement are quite different from that of film thickness measurement. Reabsorption of one dye fluorescence by the other must be avoided as it adds film thickness information to the fluorescence making it difficult to separate the temperature information contained in the fluorescence. In addition, the system must be optically thin. There are two reasons for this: (1) even if there is reabsorption (it is hard to control whether a system will have reabsorption or not, and in most practical situations reabsorption is present) an optically thin system will ensure that the reabsorption effects are minimal as the fluorescence is approximately linear with film thickness. More importantly, (2) it is easier to deal with temperature variations in the direction of observation (i.e., *x* direction in figure 3). Let us explore the last point in more detail. The goal in using fluorescence for temperature measurement is to obtain a two-dimensional map of temperature, that is, temperature variations in the plane of observation. It, however, is very likely that the temperature field also varies in the direction of observation. If this is the case and, if in particular, equation (20) holds, one can rewrite equation (6) as

$$I_f(t,T) = \int_0^t dI_f = \int_0^t I_o \exp\left[-\varepsilon(\lambda_{laser},T)Cx\right]\varepsilon(\lambda_{laser},T) \times C\Phi\, dx \qquad (22)$$

However, since $T = T(x)$, equation (22) cannot be integrated unless the temperature field as a function of x is known. This implies that, in order to correlate fluorescence to temperature, an *a priori* knowledge of the temperature field in the direction of observation is needed defeating the purpose of the technique. Thus, the two-dimensional temperature map cannot be easily inferred from the fluorescence for optically thick films if the fluorescence temperature dependence is contained in the molar absorption (or extinction) coefficient. In general, it is difficult to correlate fluorescence with temperature if there are temperature variations in the direction of observation. However, if the temperature dependence is contained in the quantum efficiency coefficient and/or the system is optically thin, the effects of temperature variation in the direction of observation on the fluorescence are not as substantial and a more accurate two-dimensional map of the temperature can be obtained. In the limit of optically thin systems, the fluorescence will correlate to the temperature at the boundary of the film, that is at location $x = 0$ for figure 3.

For optically thin systems with no reabsorption, one can use the ratiometric approach to obtain a fluorescence ratio that will correlate to temperature. Using equation (8) to calculate the fluorescence of optically thin systems, one has:

$$R(T,\lambda_{filter1},\lambda_{filter2}) = \frac{I_{f,1}}{I_{f,2}} = \frac{\varepsilon_1(\lambda_{laser},T)C_1\Phi_1\eta_1(\lambda_{filter1})}{\varepsilon_2(\lambda_{laser})C_2\Phi_2\eta_2(\lambda_{filter2})} \qquad (23)$$

and/or

$$R(T,\lambda_{filter1},\lambda_{filter2}) = \frac{I_{f,1}}{I_{f,2}} = \frac{\varepsilon_1(\lambda_{laser})C_1\Phi_1(T)\eta_1(\lambda_{filter1})}{\varepsilon_2(\lambda_{laser})C_2\Phi_2\eta_2(\lambda_{filter2})} \qquad (24)$$

The dependence of fluorescence on excitation light intensity and film thickness cancels when the ratio of the two fluorescences is used. By using this ratiometric approach on optically thin systems, temperature variations in the direction of observation are averaged over the film thickness and the fluorescence ratio can be correlated to an average temperature in the direction of observation (see figure 7).

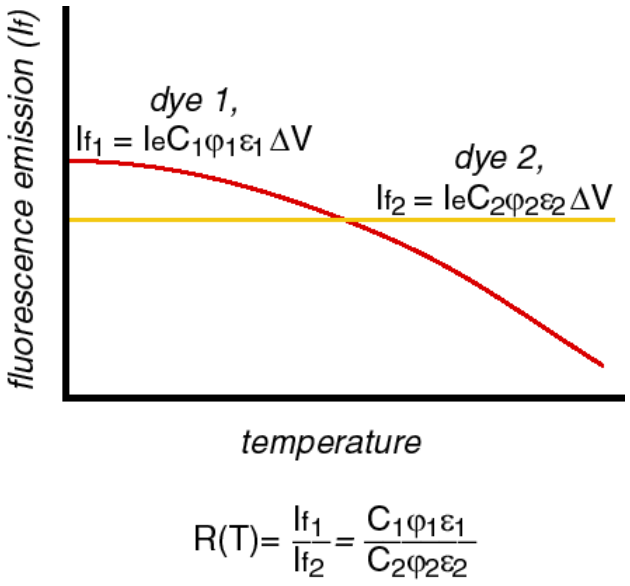


Figure 7: Temperature ratio

EXPERIMENTAL AND RESULTS

A two-12-bit-camera system was implemented in order to simultaneously capture the two fluorescence intensities (see figure 8). Dichroic mirrors and interference filters were used to separate the laser intensity from the fluorescence and the particular fluorescence emissions from each other. For oil film thickness measurements, the oil was mixed with Pyrromethene 567 and Pyrromethene 650, and for temperature measurements the combination of Pyrromethene 567 and Rhodamine 640 was implemented. Two optical flats were placed on top of each other with a 110 mm thick shim placed between them at one of the ends. This produces a linearly increasing oil film thickness.

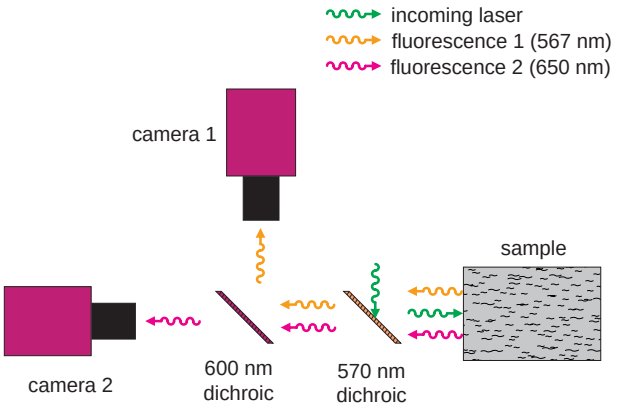


Figure 8: Experimental setup schematic

Figure 9 shows the two simultaneous fluorescence pictures (Pyrromethene 567 and Pyrromethene 650) of the oil film contained between the optical flats and their ratio. It becomes apparent how the ratiometric technique eliminates the fluorescence intensity variations due to the irregular laser intensity profile. Figure 10 depicts the change in fluorescence intensity with temperature for both Pyrromethene 567 and Rhodamine 640, and their ratio. In addition, it can be seen in Figure 11 how the ratiometric technique eliminates the laser intensity and film thickness variation, leaving only temperature information.

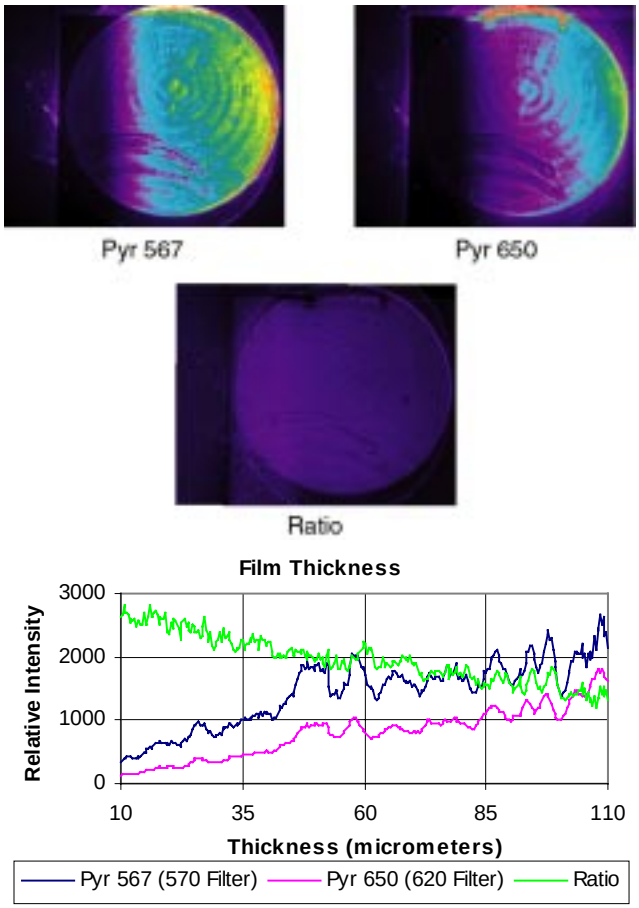


Figure 9: DELIF for oil film thickness measurement

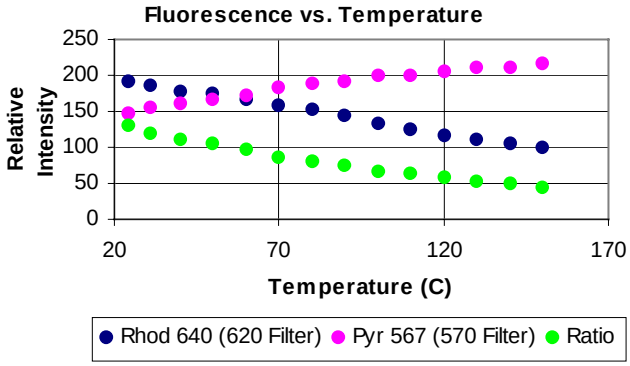


Figure 10: Fluorescence versus temperature

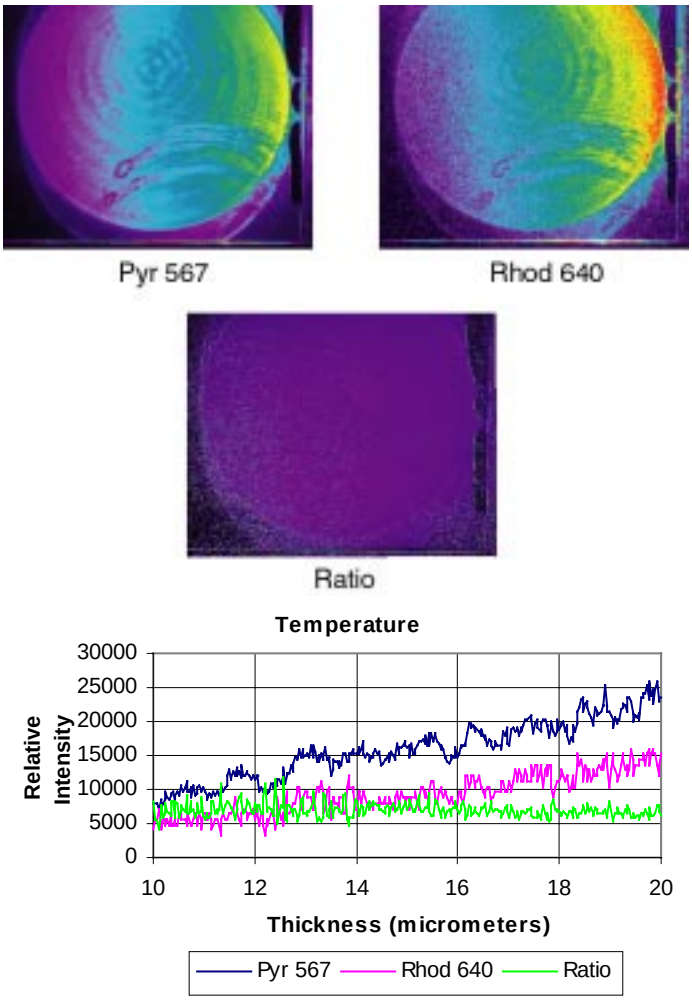


Figure 11: DELIF for temperature measurement

SUMMARY AND CONCLUSIONS

The bases for a two-dye Dual Emission Laser Induced Fluorescence (DELIF) technique for film thickness and temperature measurement were presented along with the basic equations relating these scalar measurements to dye characteristics and illumination intensity. Shown is that the non-linearity resulting from emission reabsorption, while detrimental to measurement of temperature, can be used to accurately quantify film thickness. Experimental measurements using Pyrromethene 567, Pyrromethene 650, and Rhodamine 640 demonstrate the feasibility of this technique at accurately eliminating variations in illumination intensity to extract scalar information.

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