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Emission reabsorption laser induced fluorescence (ERLIF) film thickness measurement

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Abstract

This paper presents a unique optical technique that utilizes the reabsorption and emission of two fluorescent dyes to accurately measure film thickness while minimizing errors caused by variations in illumination intensity and surface reflectivity. Combinations of dyes are selected that exhibit a high degree of emission reabsorption and each dye concentration is adjusted to create an optically thick system where emission reabsorption is intrinsic to the fluorescence of the film being measured. Film thickness information as well as excitation and dye response characteristics are all imbedded in the emission intensities of the dyes. Errors normally associated with laser induced fluorescence based film thickness measurements, including those due to optical distortion, variations in surface reflectivity and excitation non-uniformities, are minimized by observing the ratio of the dye emissions. The principle and constitutive equations characterizing emission reabsorption laser induced fluorescence (ERLIF) film thickness measurement are presented. In addition, film thickness measurements from 5 to 400 μ m with 1% accuracy are demonstrated.

Keywords: ERLIF, LIF, DELIF, film thickness

Nomenclature

- *A* area imaged onto 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 fluorescent intensity
- $dI_{f,1}$ dye 1 differential fluorescent intensity, without reabsorption
- $dI'_{f,1}$ dye 1 differential fluorescent intensity, with reabsorption
- dV differential volume element
- dx differential length in x-direction
- *F* fluorescence power
- *h* Planck's constant
- I_e exciting light intensity
- I_f total fluorescent intensity
- $I_{f,1}$ dye 1 total fluorescent intensity, without reabsorption
- $I'_{f,1}$ dye 1 total fluorescent intensity, with reabsorption
- $I_{f,2}$ dye 2 total fluorescent intensity
- I_0 exciting light intensity at x = 0

- S₀ ground state
- S₁ relaxed singlet excited state
- S'_1 singlet excited state
- t film thickness
- *x* coordinate perpendicular to plane of observation
- y coordinate parallel to plane of observation
- $\varepsilon(\lambda)$ molar absorption (extinction) coefficient at a given wavelength (absorption spectrum); effective twodye 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 strength (efficiency) at a given wavelength (emission spectrum)
- $\eta_2(\lambda)$ dye 2 relative emission strength (efficiency) at a given wavelength (emission spectrum)
- v_{EM} light wave frequency of emission
- v_{EX} light wave frequency of excitation

λ_{laser}	laser wavelength
$\lambda_{filter1}$	narrow band filter 1 wavelength
$\lambda_{filter2}$	narrow band filter 2 wavelength
τ	time

1. Introduction

Laser induced fluorescence (LIF), a technique based on the photo-excitation of a fluorophore or fluorescent dye, has gained popularity as a general-purpose visualization tool for numerous 1D, 2D and 3D applications (Ayala et al 1998, Joffe et al 1999), in particular as a flow tracer (Georgiev and Alden 1997, Kovacs 1995, Thirouard and Hart 1998). However, it has seen limited use as a quantitative tool. The reason for this stems primarily from the difficulty in separating variations in excitation illumination, surface reflectivity and optical distortion effects from fluorescent emission. Presented herein is a two-dye ratiometric technique that allows measurement of film thickness through the use of reabsorption while minimizing variations in excitation illumination, surface reflectivity and non-uniformities in optical imaging. In order to understand the principles of this technique and the advantages it presents, it is necessary to understand the fundamental characteristics of dye photo-fluorescence and how it can be used to determine a scalar of interest (1999).

As stated by Haugland (1999),

Fluorescence is the result of a three-stage process that occurs in certain molecules (generally polyaromatic hydrocarbons or heterocycles) called fluorophores or fluorescent dyes. These three stages are (figure 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 ns to 10 ns). During this time, the fluorophore undergoes conformational changes and is 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 fluorescent emission originates. Second, not all the molecules initially excited by absorption (Stage 1) return to the ground state (S_0) by fluorescent emission. Other processes such as coalitional 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 S₀. Due to



Figure 1. Fluorescence principle (Haugland 1999).

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.

From Haugland's description, it is apparent that photofluorescence can be used to characterize 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 illumination light intensity—typically from a laser. Most illumination sources are far from uniform. They vary in intensity in space and time. Pulsed lasers are particularly prone to exhibit this behaviour due to the characteristics of their resonance cavities. Use of pulsed lasers is desirable though, because of their short pulse duration (and consequently short fluorescent emission), that allow for nearly instantaneous measurements of the desired scalar.

In order to correlate two-dimensional fluorescent intensity to a scalar of interest, spatial variations in illumination intensity must be determined. This can be accomplished by using a ratiometric technique where the fluorescent intensity containing the desired scalar information is divided by the laser intensity eliminating the fluorescence dependence on excitation intensity (Inagaki et al 1997). One way to achieve this is by using two fluorescent dyes and observing the ratio of their emissions. The emissions contain the desired scalar information as well as information on the excitation light source intensity. By observing the ratio of the emissions of each dye, variations in excitation intensity can be minimized. This technique has been extensively used for measuring temperature and is known as dual emission laser induced fluorescence (DELIF) (Coppeta and Rogers 1998, Coppeta et al 1997) or two colour LIF (Sakakibara and Adrian 1999). The current work extends this technique to measure film thickness by using emission reabsorption as a means of embedding the thickness information in the fluorescence. Presented herein is the principle of emission reabsorption laser induced fluorescence (ERLIF) along with a discussion of its accuracy and limitations.



Figure 2. Fluorescence of fluid films.

2. LIF fundamentals

2.1. Optically thin versus optically thick

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

$$F = I_e \varepsilon(\lambda_{laser}) C \phi \, \mathrm{d}V. \tag{1}$$

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

- (1) excitation energy available to produce molecular transitions to higher, excited levels,
- (2) molar absorptivity (extinction coefficient), which determines how much of the incident light per molecule produces 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 fluorescent 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.

Since the length of the differential volume element is infinitesimally small, dividing equation (1) by the area A, the fluorescent intensity normal to the area A is obtained:

$$I_f = I_e \varepsilon(\lambda_{laser}) C \Phi \,\mathrm{d}x. \tag{2}$$

If the area A is assumed to be the projected area imaged onto 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 and 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 from Lambert's law of absorption (Guilbault 1990, Poll *et al* 1992), which takes into account the absorption of the excitation light by the finite fluid through which it travels:

$$I_e(x) = I_0 \exp[-\varepsilon(\lambda_{laser})Cx].$$
(3)

Consider the differential element shown at the bottom of figure 2 within a region of finite film thickness. The fluorescent intensity collected by the CCD from this fluid element is

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

Thus, from equations (3) and (4):

$$dI_f = I_0 \exp[-\varepsilon(\lambda_{laser})Cx]\varepsilon(\lambda_{laser})C\Phi \,dx.$$
 (5)

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

$$I_f(t) = \int_0^t \mathrm{d}I_f = \int_0^t I_0 \exp[-\varepsilon(\lambda_{laser})Cx]\varepsilon(\lambda_{laser})C\Phi\,\mathrm{d}x$$
(6)

such that

$$I_f(t) = I_0 \Phi\{1 - \exp[-\varepsilon(\lambda_{laser})Ct]\}.$$
 (7)

Note that the previous analysis assumes that all the fluorescent light emitted by the differential volume elements is collected by the imaging system. In reality, since the fluorescent light is directed in all directions, only a fraction of the total fluorescence emitted by a differential volume element is collected by the CCD detector. This is referred to as the 'monitoring efficiency' of the system (Wang et al 1988), and is a function of the size of the aperture of the collecting system (lens diameter in this case) and the distance from the emission location to the aperture. If the thickness over which fluorescence takes place is much smaller than the distance from fluorescence sample to collecting system aperture, the 'monitoring efficiency' can be approximated as constant (between zero and unity) over the entire thickness of the sample. Therefore, the only modification required on the analysis is a multiplication of equation (7) by this constant. This constant, as it will become clear later, is eliminated in the ratio of dye emission intensities and is, thus, inconsequential. Because of this, this constant does not appear in the following analysis.

Thus, for small values of t (thin films), equation (7) can be approximated as

$$I_f(t) \approx I_0 \varepsilon(\lambda_{laser}) C \Phi t.$$
(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 (or more properly, quasilinear) 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. Emission and absorption spectra.



Figure 4. Reabsorption schematic.

2.2. Reabsorption

Emission reabsorption is often encountered in fluorescence techniques and is generally regarded as problematic. A fluorophore behaviour is typically characterized by its absorption and emission spectrums. The absorption spectrum of a dye is a graphical representation of the molar absorptivity as a function of wavelength. In general, the absorption spectrum refers to the range of wavelengths over which excitation of the fluorophore can take place (figure 3). Similarly, the emission spectrum of a dye is the range of wavelengths over which the dye fluoresces (figure 3). When the emission spectrum of one dye overlaps the absorption spectrum of another dye (or its own absorption spectrum), reabsorption of the dye fluorescence occurs (figure 4). This has two effects: (1) it increases the fluorescent 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 fluorescent 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 fluorescent 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 at the bottom of figure 2, it is apparent that the differential fluorescent emission produced by any single element must travel back through the medium before reaching the CCD.

If there is reabsorption of the differential element fluorescence, Lambert's law must be applied to the differential fluorescent emission in order to compute the actual fluorescence collected by the CCD. Thus, assuming the situation represented in figure 4 occurs,

$$dI_{f,1} = I_0 \exp[-\varepsilon(\lambda_{laser})C_x]\varepsilon_1(\lambda_{laser})C_1\Phi_1\eta_1(\lambda) dx d\lambda \quad (9)$$

$$dI'_{f,1} = dI_{f,1} \exp[-\varepsilon_2(\lambda_{laser})C_2x] \quad (10)$$

$$dI'_{f,1} = I_0 \exp[-\varepsilon(\lambda_{laser})Cx]\varepsilon_1(\lambda_{laser})C_1\Phi_1\eta_1(\lambda) \times \exp[-\varepsilon_2(\lambda_{laser})C_2x] dx d\lambda.$$
(11)

Equation (5) has been modified in equation (9) to reflect the fact that the fluorescent 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 (Hidrovo and Hart 2000):

$$I'_{f,1}(t,\lambda_{filter1}) = \int_0^t I_0 \exp[-\varepsilon(\lambda_{laser})Cx] \\ \times \varepsilon_1(\lambda_{laser})C_1 \Phi_1 \eta_1(\lambda_{filter1}) \exp[-\varepsilon_2(\lambda_{laser})C_2x] dx$$
(12)

$$I'_{f,1}(t, \lambda_{filter1}) = \{I_0 \varepsilon_1(\lambda_{laser}) C_1 \Phi_1 \eta_1(\lambda_{filter1}) \\ \times (1 - \exp\{-[\varepsilon(\lambda_{laser}) C + \varepsilon_2(\lambda_{filter1}) C_2]t\})\} \\ \times \{\varepsilon(\lambda_{laser}) C + \varepsilon_2(\lambda_{filter1}) C_2\}^{-1}.$$
(13)

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

3. ERLIF, the reabsorption technique

3.1. Ratiometric approach

In the previous analysis, based on figure 2, and in equations (1)–(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:

$$I_0 = I_0(y, \tau).$$
 (14)

Therefore,

$$I_f = I_f(t, \lambda, y, \tau). \tag{15}$$

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

$$\frac{I_f}{I_0} \equiv R = R(t, \lambda).$$
(16)

Obtaining illumination intensity is not trivial. A twodimensional instantaneous illumination map, however, can be inferred from the fluorescence of a second dye. This is the principle behind any dual fluorescence technique:

(1) The fluorescence of dye 1 in a two-dye system contains the desired information (film thickness, temperature, or any other scalar on which fluorescence is dependent), 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 cancels resulting in a ratio that contains only the desired scalar information.

3.2. Film thickness measurement

Normally, the scalar information contained in fluorescent emission is a result of the molar absorptivity and/or quantum efficiency dependence on the scalar. This is not the case for film thickness where the fluorescence is solely a consequence of light absorption through a finite thickness for both optically thin and optically thick systems. The difference between the two optical conditions is a result of the dependence of fluorescence with film thickness. For optically thin systems, there is a quasi-linear dependence between film thickness and fluorescence. In the case of optically thick systems, there is a dependence based on the decay of light intensity as it travels through an absorbing medium. This fluorescence dependence on light intensity decay is nonlinear with film thickness, following Lambert's law.

If there is no reabsorption, dividing the two fluorescent emissions cancels the ratio dependence on thickness even for optically thick systems as both fluorescent emissions thickness dependence is governed only by the absorption of the excitation light. Consider the case of an optically thick two-dye system where each dye is excited by the same light source and there is no reabsorption. Based on equations (4)–(6), the fluorescent emission of the two dyes is given by

$$I_{f,1}(t, \lambda_{filter1}, y, \tau) = \{I_0(y, \tau)\varepsilon_1(\lambda_{laser})C_1\Phi_1\eta_1(\lambda_{filter1}) \\ \times (1 - \exp\{-[\varepsilon(\lambda_{laser})C]t\})\} \\ \times \{\varepsilon(\lambda_{laser})C\}^{-1}$$
(17)
$$I_{f,2}(t, \lambda_{filter2}, y, \tau) = \{I_0(y, \tau)\varepsilon_2(\lambda_{laser})C_2\Phi_2\eta_2(\lambda_{filter2}) \\ \times (1 - \exp\{-[\varepsilon(\lambda_{laser})C]t\})\} \\ \times \{\varepsilon(\lambda_{laser})C\}^{-1}.$$
(18)

Taking their ratio, the equation

$$R(\lambda_{filter1}, \lambda_{filter2}) = \frac{I_{f,1}}{I_{f,2}} = \frac{\varepsilon_1(\lambda_{laser})C_1\Phi_1\eta_1(\lambda_{filter1})}{\varepsilon_2(\lambda_{laser})C_2\Phi_2\eta_2(\lambda_{filter2})}$$
(19)

is obtained. Equation (19) is not dependent on t (film thickness). In order for the ratio to be thickness dependent, additional thickness information must be imbedded in one of the fluorescent dye emissions. This can be achieved by taking advantage of the reabsorption phenomena in optically thick systems where one of the fluorescent emissions is absorbed as it travels through the medium towards the CCD (figure 2). Consider the case of two dyes where the emission of dye 1 is being absorbed by dye 2 but not vice versa. As mentioned earlier, this results in a reduction in the emission of dye 1 and an increase (boosting) of the emission of dye 2. Since the external illumination intensity (laser intensity in LIF) is much greater than the fluorescent intensity of dye 1, the boosting effect can be neglected. Consequently, the total fluorescence captured by the CCD from dye 2 can still be approximated as given by equation (18). However, the reduction in total fluorescent



$$R(t) = \frac{I_{f,2}}{I_{f,1}} = \frac{\varepsilon_2 C_2 \varphi_2 (\varepsilon C + \varepsilon_2 C_2) \{1 - exp[-(\varepsilon C)t]\}}{\varepsilon_1 C_1 \varphi_1 (\varepsilon C) \{1 - exp[-(\varepsilon C + \varepsilon_2 C_2)t]\}}$$



emission from dye 1 as perceived by the CCD is quite substantial, especially if the system is optically thick and the condition $\varepsilon_2(\lambda_{filter1})C_2 \gtrsim O[\varepsilon(\lambda_{laser})C]$ is achieved. In this case, the emission thickness dependence of the fluorescence of dye 1 is governed not only by the absorption of the excitation light as it travels through the medium but also by the absorption of the fluorescent emission as it travels through the medium towards the CCD. Consequently, the total fluorescence of dye 1, as perceived by the CCD, is given by equation (13), resulting in (Hidrovo and Hart 2000):

$$I'_{f,1}(t, \lambda_{filter1}, y, \tau) = \{I_0(y, \tau)\varepsilon_1(\lambda_{laser})C_1\Phi_1\eta_1(\lambda_{filter1}) \\ \times (1 - \exp\{-[\varepsilon(\lambda_{laser})C + \varepsilon_2(\lambda_{filter1})C_2]t\})\} \\ \times \{\varepsilon(\lambda_{laser})C + \varepsilon_2(\lambda_{filter1})C_2\}^{-1}$$
(20)
$$I_{oc}(t, \lambda_{oc}, y, \tau) = \{I_o(y, \tau)\varepsilon_o(\lambda_{oc}, \tau)\}$$

$$I_{f,2}(l, \lambda_{filter2}, y, t) = \{I_0(y, t) \mathcal{E}_2(\lambda_{laser}) C_2 \Psi_2 \eta_2(\lambda_{filter2}) \times (1 - \exp\{-[\varepsilon(\lambda_{laser})C]t\})\} \{\varepsilon(\lambda_{laser})C\}^{-1}$$
(21)

$$R(t, \lambda_{filter1}, \lambda_{filter2}) = \frac{I_{f,1}^{\prime}}{I_{f,2}} = \{\varepsilon_1(\lambda_{laser})C_1\Phi_1\eta_1(\lambda_{filter1}) \\ \times \varepsilon(\lambda_{laser})C(1 - \exp\{-[\varepsilon(\lambda_{laser})C + \varepsilon_2(\lambda_{filter1})C_2]t\})\} \\ \times \{\varepsilon_2(\lambda_{laser})C_2\Phi_2\eta_2(\lambda_{filter2})[\varepsilon(\lambda_{laser})C + \varepsilon_2(\lambda_{filter1})C_2] \\ \times (1 - \exp\{-[\varepsilon(\lambda_{laser})C]t\})\}^{-1}.$$
(22)

The excitation light intensity dependence is cancelled by taking the ratio of the emission of the two dyes, leaving a



Figure 7. Calibration fixture.

quantity that is only dependent on film thickness. Since the 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 (figure 5).

4. Experimental validation

4.1. Experimental set-up

In order to demonstrate the feasibility of the ERLIF technique, a two-camera system was developed (figure 6). The system consists of two 12-bit Princeton Instrument CCD cameras mounted on a single lens, optical-path-splitting module. The use of a single lens simplifies alignment and minimizes distortion between the two cameras. Two dichroic mirrors are used in order to separate the laser and fluorescent emissions. The first dichroic is located outside the module and is used to simultaneously steer and separate the laser from the fluorescent emissions. The second dichroic is located inside the module and serves two functions: (1) to separate and (2) to steer the low and high wavelength fluorescent emissions. Interference filters are used just before the CCD cameras in order to isolate a particular wavelength emission. The module also contains adjustment optics that allow for mechanical alignment of the two images captured by the respective CCD cameras. Further alignment is accomplished by computer processing of the two images. A piece of sandpaper is placed in front of the



Figure 8. Laser beam profile.



Figure 9. Comparison of the film thickness LIF signal for Pyrromethene 567 and Pyrromethene 650 versus their ratio (optically thick system with strong reabsorption).

module and imaged by both CCD cameras. A cross correlation particle image velocimetry (PIV) algorithm (Hart 1999) is then used to locally correlate the two images in order to find the displacement vectors (at the sub-pixel level) for each pixel position, which will subsequently be used on all other images when computing the ratio.

The two dyes selected to demonstrate the feasibility of the technique were Pyrromethene 567 and Pyrromethene 650. These dyes were selected because (1) both are excitable by the 532 nm line of the Nd:YAG, (2) wide separation exists between the primary emission peak of each dye, (3) both dyes have high quantum efficiencies, (4) the emission peak of Pyrromethene 567 is highly absorbed by Pyrromethene 650 and (5) both are non-hazardous.

A calibration fixture was fabricated in order to provide a linearly increasing film thickness against which the technique could be tested. The fixture consisted of a quartz optical flat that formed the top and a quartz flat set at an angle with an



Figure 10. Film thickness LIF signal for Pyrromethene 567, Rhodamine 640 and their ratio (optically thin system with negligible reabsorption).



Figure 11. Film thickness LIF signal for Pyrromethene 567 under low and high back surface reflectivity conditions.

inside reservoir channel etched around it (figure 7). When joined together, the optical flats produced a linearly increasing gap that was filled with liquid in order to produce a known film thickness. The fixture was measured using a CMM to verify the thickness of the gap within the calibration area.

4.2. Experimental results

Figure 8 depicts the laser beam intensity profile. Strong spatial intensity variations can be observed with fluctuations of as much as 400% between the highest and lowest intensity regions of the beam. Since the technique was originally



Figure 12. Film thickness fluorescence ratio signal under low and high back surface reflectivity conditions.





Pyrromethene 650

Figure 13. LIF pictures of an oil film deposited on top of a US mint quarter coin.

developed for lubrication and tribological purposes, the system was initially tested on its ability to measure oil films. A dye concentration of 8×10^{-4} mol 1^{-1} of oil was used for both dyes. Figure 9 shows the fluorescence dependence on film thickness for Pyrromethene 567 and Pyrromethene 650. There is a noticeable increase in fluorescence with film thickness that

becomes nonlinear as the film thickness increases (optically thick system). It is apparent, however, that the laser intensity fluctuations are embedded within the film thickness information, making it difficult to separate the two. The bottom of figure 9 shows the ratio of the two fluorescent emissions. Note the disappearance of the laser intensity fluctuations,





Figure 14. Surface topography of a US mint quarter coin obtained by the ERLIF method.

illustrating that the laser intensity information is cancelled in the ratio. Note also that the ratio has a nearly linear dependence on film thickness. As explained earlier, this is a consequence of reabsorption within the optically thick system. For the dye combination used, it was possible to resolve film thickness from 5 to 400 μ m with 1% accuracy. Accuracy can be greatly improved over a specific range of film thickness through careful dye selection and adjustment of dye concentrations.

As further evidence of this reabsorption technique for measuring film thickness, the tests were repeated on an optically thin system. A dye combination consisting of 8 \times 10^{-5} mol l⁻¹ Pyrromethene 567 and 2.4×10^{-4} mol l⁻¹ Rhodamine 640 dissolved in oil was used. Although this twodye system exhibits reabsorption (since the emission spectrum of Rhodamine 640 overlaps the absorption spectrum of Pyrromethene 567), the low concentrations used for both dyes and the fact that only thickness up to 45 μ m were considered results in an optically thin behaviour where reabsorption is minimal. Consequently, the fluorescence dependence of both dyes over the film thickness range of the calibration fixture is linear. This results in a constant value for the ratio over the thickness range. Figure 10 shows the fluorescence dependence on film thickness for Pyrromethene 567 and Rhodamine 640. Again, a noticeable increase in fluorescence with film thickness is observed for both dyes, and laser intensity fluctuations are evident within the film thickness information. The bottom of figure 10 shows the ratio of the two emissions. As before, the laser intensity fluctuations disappear with the ratio. However, in this case, the ratio remains constant over the film thickness range. There is no dependence of the ratio on film thickness because the system is optically thin and reabsorption has minimal influence. While an optically thin condition cannot be used for the measurement of film thickness, it is appropriate when a ratiometric approach is to be used to measure scalar quantities such as temperature, pH and pressure (Coppeta and Rogers 1998).

Because the ratio of the two dye emissions in the ER-LIF technique is independent of illumination intensity, it is also independent of surface reflectivity characteristics and nonuniformities due to factors such as optical distortion. For example, a LIF signal for a film is dependent on the optical characteristics of the materials surrounding it. In a typical system, this would usually be the reflectivity and transmissivity of the window of observation and the reflectivity of the back surface. Even if a known uniform excitation source were to be used, an *in situ* calibration would be required for such a measurement to obtain accurate results. This is rarely feasible to any reasonable accuracy. With the use of ERLIF, a very precise external calibration system can be used, as it is independent of illumination and imaging non-uniformities. Figure 11 shows the LIF signal of Pyrromethene 567 for two different back surface reflectivities. The quartz calibration fixture was placed on top of two different materials. The first material was a matt black rubber piece, while the second material was a polished steel block, termed low reflectivity material and high reflectivity material, respectively. It can be seen that for the high reflectivity material the LIF signal is around twice that of the low reflectivity material. Figure 12 shows the ERLIF ratio signal for the same conditions. It can be seen that there is no measurable difference in the ratio signal despite the different reflectivities of the materials used as backing.

As a tangible example of the accuracy of the ERLIF technique, it was used to infer the surface topography of a US mint 25¢ coin. The coin was placed flat in the bottom of a beaker and a small layer of oil premixed with Pyrromethene 567 and Pyrromethene 650 was deposited on top of the coin. ERLIF was then used to measure the film thickness of the oil on top of the coin. Due to the topography of the coin, the thickness of the oil film on top of the quarter generates a female match of the coin features: raised surface regions produce thin film thickness (low fluorescence) and vice versa-surface depressions produce thick film thickness (high fluorescence). Figure 13 shows the Pyrromethene 567 and Pyrromethene 650 fluorescence from the oil film lying on top of the coin. It is impossible to infer coin surface features from the Pyrromethene 567 fluorescence; nonetheless, this fluorescence provides a reasonable map of the laser beam intensity profile. Some of the coin features are apparent in the Pyrromethene 650 fluorescence but, again, the fluctuations in laser intensity profile prevent a clear interpretation of the coin features from the basic LIF signal. Figure 14 shows the ratio of the two fluorescence signals. The surface topography of the quarter becomes evident-not simply as an image but as a quantifiable topography of the coin surface. The clarity of this figure is evidence of the ability of this technique to eliminate local illumination variations, while preserving film thickness information.

5. Summary and conclusions

The bases for a two-dye emission reabsorption laser induced fluorescence (ERLIF) technique for film thickness measurements have been presented. The core of the technique relies on the use of a ratiometric approach for the purpose of suppressing excitation intensity information from the fluorescent emission. The basic principles and equations behind photofluorescence, optically thin and thick systems and reabsorption were introduced in light of the technique and in order to develop a theoretical framework on which to base the technique. It is shown that the nonlinearity resulting from emission reabsorption, while detrimental to the measurement of most scalars, can be used to accurately quantify film thickness when a ratiometric approach is used. Implementation of the technique using a two-camera system and a calibration fixture demonstrates the feasibility of this unique technique and its ability to accurately determine film thickness despite variations in illumination intensity and surface reflectivity.

The technique is limited in the range of film thickness that can be measured. The thickness range and resolution depends, among other factors, on the optical thickness of the system and the level of reabsorption that can be achieved. The current work demonstrates film thickness measurements in the 5–400 μ m range with 1% accuracy.

The ERLIF technique proves useful in that it minimizes errors caused by variations in excitation intensity. It allows for calibration without the need to replicate the same optical characteristics of the system being studied. Furthermore, it allows the use of a single calibration, for a specific dye combination, that can be used on a number of different experiments regardless of the optical characteristics of the system components involved. It allows for instantaneous 2D mapping of film thickness-useful when studying transient phenomena. The ERLIF technique was originally developed to measure oil film thickness in tribological systems but its application can be extended to the measurement of other film compositions. Measurement of free boundary films will especially benefit from the use of the technique, where techniques such as interferometry are not easily implemented.

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