Application of 2-D LIF temperature measurements in water using a Nd : YAG laser

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Abstract The application of Laser Induced Fluorescence (LIF) for temperature measurements in water using a Nd : YAG laser is investigated. A natural convection problem is used to test the applicability of LIF in the temperature range of 20–60°C. The measured temperature field is compared with numerical results and the influences of shadowgraph effects on the measured temperature field are investigated. An accuracy of 1.7°C is attained if shadowgraph effects can be neglected. This only holds if correction for photobleaching and variation of laser power output is applied.

List of symbols

- $I$: fluorescence intensity
- $I_0$: excitation intensity
- $C$: rhodamine B concentration
- $\varepsilon$: molar absorptivity
- $\varepsilon_{\text{max}}$: maximum molar absorptivity
- $I_{\text{max},f}$: maximum emission
- $A$: fraction of collected light
- $\Phi$: quantum yield
- $\lambda_e$: wave length of the excitation light
- $\lambda_f$: wave length of the fluorescence light
- $D$: depth of the test section
- $H$: height of the test section
- $B$: width of the test section
- $I_n$: normalized intensity
- $I_{T,\text{low}}$: fluorescence intensity at the lowest reference temperature
- $I_{T,\text{high}}$: fluorescence intensity at the highest reference temperature
- $I_{\text{max}}$: maximum intensity
- $I_0$: corrected excitation intensity
- $l$: averaging length in a frame
- $x_1$: start test section
- $x_2$: end test section
- $n$: refractive index
- $T$: local temperature
- $\Delta I$: variation in intensity due to shadowgraph effects

1 Introduction

Although velocities can be measured quite easily in a 2-D plane by using PTV and PIV, whole field or 2-D temperature measurement techniques are less developed and used. At this moment the application of non-intrusive temperature measurement techniques is limited. The major limits are temperature range, accuracy and gradients. The most common technique is based on liquid crystals. Several researchers have reported quantitative measurements using liquid crystals, Dabiri and Gharib (1990, 1991) and Sillekens (1995). The accuracy relative to the temperature range is, however, not large. A typical accuracy according to Sillekens (1995) is approximately 0.6°C for 6°C temperature range.

A less common technique is based on Laser-Induced Fluorescence (LIF) which uses temperature sensitive fluorescent dyes excited by laser light, Sakakibara et al. (1993, 1997) and Nakajima et al. (1990). A great advantage of LIF above liquid crystals is the better accuracy relative to the temperature range which is typically 1.5°C for a 40°C temperature range, Sakakibara and Adrian (1997). LIF is based on natural fluorescence of molecules and atoms which is induced by absorption of a photon (energy). This absorption causes a transition from the ground state to an excited state. Part of the absorbed energy is re-emitted during spontaneous transition from the excited state to the ground state (fluorescence). The fluorescence occurs at a longer wavelength than absorption due to energy loss in the excited state and the fluorescence intensity $I$ can be described by the following equation, Berlman 1971; Bindhu et al. 1996:

$$I = I_0 A \Phi e C \frac{\lambda_e}{\lambda_f}$$  \hspace{1cm} (1)

where $I_0$ is the intensity of the excitation beam, $C$ the concentration of the fluorescent dye, $e$ the molar absorptivity, $A$ the fraction of the available light collected and $\Phi$ the quantum yield. The wavelength ratio $\lambda_e/\lambda_f$ accounts for the energy loss in the excited state. The quantum yield turns out to be temperature dependent, resulting in a temperature dependent fluorescence intensity. The observed temperature effect on the quantum yield is possibly induced by quenching effects. For increasing temperature the quenching effect increases resulting in a decrease of the quantum yield. This dependence makes LIF useful for temperature measurements.
Previous reports on temperature measurements all used a relative stable CW Ar⁺ laser for the excitation source which guarantees a rather constant intensity \( I_0 \). In this work, however, a Nd:YAG laser is used (Spectra-Physics GCR-150) operating at 532 nm. This laser is chosen for its high power output which makes it possible to examine larger flow field areas. The laser produces a 6 ns pulse containing 200 mJ at a repetition frequency of 30 Hz. Due to this short pulse duration it is also possible to acquire instantaneous images with a high signal to noise ratio (Karasso and Mungal 1997).

This work focuses on the applicability and on the estimation of temperature measurement errors using the fluorescent dye rhodamine B in combination with a Nd:YAG laser. Problems arising due to the use of a Nd:YAG laser are discussed within the next section and an experimental procedure accounting for these problems is described. This procedure is used to measure the temperature field of a natural convection problem. The measured temperature field is compared with a calculated temperature field. From this comparison an estimate of the overall accuracy is derived. The sources which are responsible for this error are examined thoroughly in the last section.

2 Experimental method and procedure

2.1 Fluorescent dye

Previous research identified rhodamine B as a temperature sensitive fluorescent dye. A decrease of fluorescence intensity of 2–3% per Kelvin is not unusual according to Nakajima et al. (1990) and Sakakibara et al. (1993). The decrease of fluorescence intensity for increasing temperatures is caused by a decrease in quantum yield \( \phi \) according to López Arbeloa et al. (1986, 1991). This decrease is not wavelength dependent as can be concluded from the fluorescence spectra shown in Fig. 1 which are obtained by using an Ar⁺ laser and a spectrograph (SpectraPro 275). As one can see a higher temperature results in an overall decrease of the emission spectrum. This feature combined with the fact that fluorescence occurs at a longer wavelength than absorption makes it possible to separate the fluorescence signal from elastically scattered laser light (without energy loss). This is achieved by placing a long-wave pass filter (Schott Glass OG550) in front of the camera lens which blocks light with a wavelength smaller than 550 nm almost completely.

The absorption spectrum maximum of rhodamine B dissolved in water is near 550 nm according to López Arbeloa and Ruiz Ojeda (1981) (Fig. 1). From this figure it can be concluded that the Nd:YAG laser is suitable for excitation. Besides, the excitation wavelength of this Nd:YAG laser (532 nm) is closer to the absorption maximum than the excitation wavelength of an Ar⁺ laser (514.5 nm). This means that for an equal excitation intensity a Nd:YAG laser will result in a higher fluorescence intensity (Eq. 1).

In this work the fluorescent dye rhodamine B (MW = 379.02) is dissolved in 5 μm filtered demineralized water with a concentration of typically \( 10^{-7} \) M. Demineralization of the used water is necessary because non fluorescent tracers can act as quenchers according to Nash et al. (1995).
However, near the symmetry plane the flow can be considered which results in a velocity component in the third dimension. This laminar natural convection problem is usually a 3-D flow according to Henkes (1990) and Le Que (1985). Therefore the measured excitation intensity which is used for the temperature calculation has a total accuracy of about 0.8% (0.55% for the pulse to pulse variation and 0.25% for the long-term variation).

The test section used to create the natural convection flow is shown in Fig. 3. This test section consists of a closed cavity bounded by two aluminum heat exchangers, which form the two vertical walls. The horizontal walls of the cavity, as well as the front and back lateral walls are made of 15 mm thick glass. The natural convection flow in the cavity is induced by heating the left vertical wall at 58.8°C and cooling the right vertical wall at 14.6°C. This results in a Grashof number of $4.5 \times 10^7$ (properties taken at the mean temperature of 36.7°C). This Grashof number will result in a laminar flow according to those relations found in Le Que and Alziary de Roquefort (1985). This laminar natural convection problem is usually a 3-D flow problem due to energy loss at the front and rear of the cavity which results in a velocity component in the third dimension. However, near the symmetry plane the flow can be considered 2-D according to Mallinson and de Vahl Davis (1977) if $D/B > 1.2$ and $H/B > 1.2$, which is valid for this test section ($D/B = 5$, $H/B = 2$).

### 2.3 Calibration

In order to measure temperatures a calibration curve between temperature and fluorescence intensity is necessary. To obtain this calibration curve the temperature dependence of the fluorescence intensity is measured by heating the cavity (Fig. 3) to uniform temperatures. The uniform temperatures are achieved by heating both vertical walls at the same temperature. The temperature dependence is measured in the middle of the cavity in five different experiments resulting in 45 measurement points (Fig. 4). This figure shows a S-shaped decrease of the fluorescence intensity as was expected by Nakajima et al. (1990). Because of this S-shape the calibration curve is approximated by a third order least square method resulting in an averaged random error over the whole temperature range of 1.7°C (95% confidence).

The presented intensities in Fig. 4 are normalized. This normalization is done because the fluorescence intensity for a certain temperature varies between different experiments. This variation is mainly caused by variations in dye concentration ($C$ in Eq. (1)) and the relative position of the test section in the experimental set up. A variation of the position of the test section in example in positive $x$-direction (Fig. 2) results in a decrease of the excitation intensity ($I_e$ in Eq. (1)), whereas a variation of the position in $z$-direction results in a variation of the fraction of the available light collected by the camera ($A$ in Eq. (1)). The normalization is carried out by measuring the fluorescence intensity field for two constant temperatures, a high (55–60°C) and a low (20–25°C) temperature. This results in the following normalized fluorescence intensity field:

$$I_n(x,y) = \frac{I(x,y) - I_{T,high}(x,y)}{I_{T,low}(x,y) - I_{T,high}(x,y)}$$

where $I_{T,low}$ is the local measured fluorescence intensity at the low normalization temperature $T_{low}$, $I_{T,high}$ is the fluorescence intensity at the high normalization temperature $T_{high}$, and $I$ is the measured local intensity in the considered fluid flow. The

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**Fig. 3.** Test section, $H = 50$ mm, $B = 25$ mm and $D = 120$ mm

**Fig. 4.** Relation between the fluorescence intensity and temperature, ---: third-order approximation
The basic idea is that the dependency of the temperature measurement on the measuring device, concentration and the camera position need to be canceled out. Therefore, before every experiment two raw uncorrected reference intensity fields are recorded at a high temperature ($I_{T,\text{high}}$) and a low temperature ($I_{T,\text{low}}$). These images contain the information of the above described parameters. The influence of these parameters will finally cancel out the moment the measured intensity fields are divided by these reference intensity fields (Eq. (2)). Also the nonuniformity which occurs in all fields will cancel out by this procedure.

The actual temperature is now obtained by calculating the normalized local fluorescence intensity using the intensity field of the observed flow field and the intensity fields at the normalization temperatures. In this work the normalization temperatures are 19.8 and 55.2°C. Then, the third-order approximation and the calculated normalized fluorescence intensity are used to obtain the temperature field.

### 3 Temperature measurement in a cavity

The temperature field of the natural convection flow is measured 20 h after starting heating and cooling. This is long enough for the considered flow problem to evolve to a stationary flow. The measured temperature field shows a temperature stratification in the middle of the cavity (Fig. 5). Along a horizontal line through the test section starting at the hot wall the measured temperature field shows a temperature decrease followed by a remarkable increase. In the middle the temperature along this line is relatively constant. At the cold side the temperature first slightly increases and finally drops as one reaches the well.

In order to verify the experimental measured temperature field, the temperature field is also calculated using the finite element package SEPRAN (Segal 1984). This package was extensively validated in an earlier study by Sillekens et al. (1998). In this study the number of elements used was 40 in horizontal and 20 in vertical direction. As boundary conditions for the velocity at all walls the no slip conditions are used. For the temperature the top and bottom wall are assumed adiabatic and the vertical walls isothermal. Due to the relative large temperature difference in the cavity (44.2°C) it is necessary to assume the thermal expansion coefficient and the viscosity temperature dependent (Gray and Giorgini 1976). The temperature field shown in Fig. 5 is therefore calculated with temperature dependent viscosity and thermal expansion coefficient.

A direct comparison shows in both temperature fields a similar temperature stratification in the middle of the cavity. However, near the vertical walls the temperature fields are quite different. In contrast to the measured temperature field the calculated temperature field shows a more realistic profile, the temperature increases towards the hot wall and decreases towards the cold wall. A more detailed comparison will be made using a vertical cross-section at $x/B = 0.5$ shown in Fig. 6, and a horizontal cross-section at $y/H = 0.5$ shown in Fig. 7. In these figures the solid line represents the calculated temperature profile. The experimental measured temperatures are

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**Fig. 5a** Measured temperature field; **b** calculated temperature field

**Fig. 6.** Vertical cross section at $x/B = 0.5$, $-$: calculated; $\times$: third-order approximation

**Fig. 7.** Horizontal cross section at $y/H = 0.5$, $-$: calculated; $\times$: third-order approximation; $+$: corrected for shadowgraph effects
represented by the cross marks (×). From Fig. 6 it can be concluded that the measured temperature is within 1.7 °C of the calculated temperature profile. Only one point at y/H = 0.2 seems to have a rather large error (2.5 °C). This large error is probably caused by a non-uniformity in the background due to the presence of a thermocouple. From Fig. 7 it can be concluded that in the center the measured temperature is relatively close to the calculated temperature. However, near the vertical walls the difference between experimental and numerical is at maximum 150 °C. This large temperature difference near the vertical walls is due to the shadowgraph effects, as will be described in the next section.

4 Error analysis

4.1 Measurement errors
The final error in the measured temperature depends mainly on the error in the calibration function. The error in the calibration function introduces the error in the temperature field by transforming the intensities to temperatures. The accuracy of the measured temperature using the calibration function is not constant throughout the measurement field and depends major on photobleaching, stability of laser power output, pH, self-absorption and shadowgraph effects.

An important factor influencing the accuracy of the measurement is the degradation of the dye which is introduced by the high laser power output. This degradation causes the fluorescence intensity to decrease during excitation. This decrease occurs due to chemical/photochemical decomposition and is also called photobleaching (Saylor 1995). To minimize photobleaching the excitation time is minimized during the experiments. Because it takes the laser about 30–35 s to stabilize, the maximum excitation time during one measurement is therefore chosen to be 40 s. During one experiment at least three measurements are necessary, two reference fields for the normalization and one intensity field of the considered temperature field. This means that the dye is exposed at least 120 s to the high excitation power of the laser. In order to correct for the occurring photobleaching, the average decrease of the fluorescence intensity in the cavity due to photobleaching is examined at constant temperature. The intensity decrease during this experiment is presented in Fig. 8. From this figure it can be concluded that the fluorescence intensity decreases about 2–3%. Because of this large error it is necessary to correct for photobleaching as is done in the temperature field shown in Fig. 5. This correction is applied by dividing the measured intensity fields by a second order approximation of the intensity ratio \( I/I_{\text{ref}} \) shown in Fig. 8. Using this second order approximation the uncertainty of this correction is about 0.4 °C.

The correction procedure for variations in laser power output described in Section 2.2 is also applied in Fig. 5. The uncertainty of this correction is about 0.25%, and therefore results in a random error in the measured temperature. The error due to variation in laser power output after correction is at maximum 0.1 °C. The influence of pH on the measured temperature is also investigated and it turned out to be of minor influence on the fluorescence intensity for pH 5 to 12.

From Fig. 1 it can be seen that an overlap exists between the absorption and emission spectrum. This overlap results in a partial absorption of the fluorescence signal also called self-absorption, which depends on the local dye concentration according to Lemoine et al. (1996). Part of this absorbed fluorescence signal is re-emitted. This re-emission is, however, temperature dependent due to the temperature dependent quantum yield. This will lead to an increasing self-absorption for increasing temperature and will therefore influence the obtained intensity field. For the used concentration (10⁻⁷ M) and the small test section this effect can, however, be neglected.

The errors described in this section account for an uncertainty of about 0.5 °C. However, the uncertainty of the measurements is about 1.7 °C, meaning an uncertainty of 1.2 °C is still unexplained. The most likely causes for this uncertainty are random noise in the CCD camera and the frame grabber and concentration variations of non fluorescent tracers in water (Section 2.1).

4.2 Shadowgraph effects
The result of shadowgraph effects is a local variation of the excitation intensity, which are caused by in homogeneous variation of the refractive index inside the considered fluid flow. This variation of the refractive index is caused by a variation in density. The density is finally dependent on the temperature which makes the refractive index temperature dependent. It turns out that shadowgraph effects are very sensitive for temperature gradients, therefore the fluorescence intensity is no longer only dependent on the water temperature but also on the local temperature gradient. The variation of the excitation intensity which occurs due to the variation in refractive index \( \Delta I/I \) can be approximated using the following equation (Merzkirch 1974):

\[
\frac{\Delta I}{I} = \frac{I_0(y,z) - I_0^*(y,z)}{I_0^*(y,z)} = I \int_{x = x_i}^{x = x_f} \left( \frac{\partial^2}{\partial y^2} + \frac{\partial^2}{\partial z^2} \right) \ln n(x,y,z) \, dx
\]

where \( I_0 \) is the excitation intensity without shadowgraph effects, \( I_0^* \) the calculated excitation intensity, \( x_i \) the start of the frame, \( x_f \) the end of the frame and \( I \) the averaging length in one
frame. The thermal sensitivity of the refractive index in water (with \( T \) the temperature in °C) can be written as (Washburn 1930)

\[
n(T) = n(\lambda_c, 20°C) - 10^{-5}(0.124(T - 20) + 0.1993(T^2 - 400) - 5E(-5)(T^4 - 1.6E4))
\]

\[
n(\lambda_c, 20°C) = \sqrt{1.76148 - 0.013414\lambda_d + 0.0065438/(\lambda_d - 0.11512)^2}
\]

with \( \lambda_d = (\lambda_c \times 10^6)^2 \)

Using Eqs. (3)–(5) (Nd : YAG laser \( \lambda_c = 532 \times 10^{-9} \text{m} \)) and applying these to the temperature field shown in Fig. 5a the local variations due to shadowgraph effects are calculated. The resulting relative variation of the excitation intensity in the light sheet is shown in Fig. 9. In this figure the light sheet enters the cavity at \( y/H = 0 \) with uniform intensity and leaves the cavity at \( y/H = 1 \). The hot vertical wall is located at \( x/B = 0 \) and the cold wall at \( x/B = 1 \). From this figure it can be concluded that near the vertical walls the variation due to shadowgraph effects is large. Near the hot vertical wall the light is refracted from the wall as expected by the decreasing refractive index for increasing temperature. The same holds for the cold wall, here the light refracts to the cold wall.

The local variation of excitation intensity at \( y/H = 0.5 \) is shown in Fig. 10. From this figure the large difference between experimental and numerical results in Fig. 7 can be explained. Near the hot wall the excitation intensity decreases about 11%. Due to this decrease the measured temperature using LIF will increase with about 45°C. The increase of excitation intensity at \( x/B = 0.05 \) leads to a decrease in the measured temperature as visible in Fig. 8. Near the cold wall the opposite happens. The excitation intensity increases locally approximately 3.7% and the excitation intensity at \( x/B = 0.95 \) decreases, resulting in an increased temperature of about 15°C. It turned out that a quantitative correction is only partly possible using Eq. (3). This can be concluded from Fig. 7 in which the cross marks (+) represent the calculated temperatures after correction for shadowgraph effects. The still existing differences between numerical and experimental results after correction are caused by two severe simplifying assumptions made to obtain Eq. (3).

The first assumption is that each light ray undergoes only infinitesimal or even negligible deviations inside the inhomogeneous temperature field, but that the ray has a non-negligible curvature (Merzkirch 1974). This assumption is justified in case of a very narrow test section. In our set up this is the case, but the temperature gradients are too large to justify this assumption and therefore a calculated intensity decrease will be smaller than the experimental intensity decrease. The second assumption is a linearization of the used mapping function (Merzkirch 1974). This linearization is, however, a very radical simplification since displacements can become large for larger temperature gradients as is the case in the considered temperature field. This second simplification also results in a calculated intensity field with smaller intensity variations. Beside these simplifications local photobleaching results in a non-linear relation between the fluorescence intensity and the excitation intensity.

5 Conclusion

It is demonstrated that non-intrusive temperature measurements based on Laser-Induced Fluorescence are possible using the Nd : YAG laser. This is done by applying the measurement technique to a natural convection flow in a cavity. The flow is induced by heating one vertical wall to 58.8°C and the other vertical wall to 14.6°C resulting in a Grashof number of \( 4.5 \times 10^7 \). The uncertainty is at maximum 1.7°C as can be concluded from a comparison of the experimental temperature field with the numerically obtained temperature field and the calibration curve between temperature and fluorescence intensity. The calibration curve decreases according a S-shaped curve. This S-shaped curve was expected by Nakajima et al. (1990), but not experimentally obtained. Nakajima et al. (1990) obtained a curve of second degree in the temperature range 20 to 70°C using an Ar⁺ laser, as did Sakakibara et al. (1993, 1997). Because of this S-shaped calibration curve the fluorescence intensity is approximated by a third-order least-squares method. Due to intensity variations between different experiments it is necessary to normalize this calibration function every experiment. This normalization is carried out by using...
two reference intensity fields at different temperatures. Previous reports on temperature measurement, (Sakakibara et al. 1993, 1997; Nakajima et al. 1990), used only one reference intensity field. In this study it is found that one reference intensity field in the temperature range 20–25°C is not sufficient and leads to large errors for temperatures higher than 40°C. At least two reference fields are necessary to obtain a measurement uncertainty of 1.7°C at maximum. Beside calibration using two normalization fields, corrections must be made for photobleaching and laser instability.

The most important restriction for the application of LIF is the occurrence of shadowgraph effects. A temperature field with large temperature gradients will lead to variations in excitation intensity due to the temperature dependent refractive index. As a rule of thumb one can say that the error due to shadowgraph effect remain below 1°C if the temperature gradient does not exceed 1°C/mm.

In the present study this variation in excitation intensity is calculated using the numerically obtained temperature field. A quantitative temperature correction is not possible using the described method due to too severe simplifications and photobleaching. This photobleaching effect results in a non-linear relation between excitation and fluorescence intensity.

The present technique can be improved by avoiding averaging, avoiding shadowgraph effects and minimizing photobleaching effects. The averaging procedure can be avoided by measuring the pulse energy. If one measures pulse energy a temperature field can be measured every 1/30 s. The second improvement is based on minimization of photobleaching by decreasing the excitation time. This decrease of excitation time can be achieved by using a beam dump during the stabilization period of the laser (35 s) which is removed during the actual measurement. The influence of shadowgraph effects can locally be minimized by a smart orientation of the light sheet. Shadowgraph effects are minimized when the light sheet propagation direction is perpendicular to the temperature gradients in the temperature field.

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