A Novel Technique for Quantitative Temperature Mapping in Liquid by Measuring the Lifetime of Laser Induced Phosphorescence

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Abstract : A unique method is described in the present paper for quantitative mapping of temperature field in a liquid flow by taking advantage of the phosphorescence lifetime dependence on temperature. This methodology represents an extension of the Molecular Tagging Velocimetry (MTV) technique. As in MTV measurements, the phosphorescence signal is imaged at two successive times within the lifetime of the tracer. The phosphorescence lifetime, estimated from the intensity ratio of the two images at each point, is calibrated with respect to the temperature. This procedure eliminates the effects of the variation of the incident illumination. The method described in the present paper also allows the simultaneous quantification of velocity and temperature fields in a fluid flow by using the same phosphorescent tracer (1-BrNp·Gβ-CD·ROH) and the same optical and equipment setup. While the displacements of the tagged regions between two interrogations provide the estimate of fluid velocity vectors, the intensity ratio of the two images represents the fluid temperature distribution. The implementation and application of the new technique are demonstrated by conducting simultaneous temperature and velocity measurements in a pulsed cold jet discharging into hot ambient fluid.

Keywords : Temperature measurement, Molecular tagging velocimetry, Simultaneous velocity and temperature measurement, Phosphorescence lifetime measurement.

1. Introduction

It is well known that both fluorescence and phosphorescence are molecular photoluminescence phenomena. Compared with fluorescence, which typically has a lifetime on the order of nanoseconds, phosphorescence usually has a much longer lifetime and can last anywhere from microseconds to minutes. Since the emission intensity is a function of the temperature of the molecule for some substances, both the fluorescence and phosphorescence of tracer molecules in a fluid flow may be used for temperature measurements by quantification of photoluminescence intensity.

Laser Induced Fluorescence (LIF) technique has been widely used for temperature measurements of fluid flows in recent years. For dilute solutions and low excitation energies, the LIF

intensity is linearly proportional to the exciting light intensity, the concentration of the fluorescent dye and the quantum efficiency of the fluorescent molecule. For some dyes, such as Rhodamine B, the quantum efficiency is a function of temperature. Therefore, the fluorescence intensity may be considered, in principle, to depend only on temperature if the excitation laser intensity is uniform and the fluorescent dye concentration remains constant in the measurement region. However, in practice, it is very difficult, if not impossible, to ensure a non-varying laser illumination intensity distribution, especially in flows with index of refraction variations. This may cause significant error in the measurement results.

In order to overcome this problem, intensity-ratio-based LIF techniques have been proposed recently, which include the two-dye LIF technique (Coppeta and Roger, 1998; Sakakibara and Adrian, 1999) and the two-emission-band LIF technique (Lavieille et al., 2001). For these intensity-ratio-based LIF techniques at least two cameras with various optical filters are required for the temperature mesurements in unsteady flows. Therefore, a very careful image registration or coordinate mapping procedure has to be conducetd during the experiment in order to get the quantitative spatial relation between the two images. In addition, other complications need to be carefully considered, such as the spectral conflicts and photobleaching behavior of the two dyes (Coppeta and Roger, 1998). Since the LIF signal in a narrow spectrum band is always very weak, issues like signal-to-noise ratio and the choices of the image recording system and optical filters, need to be carefully considered for the two-emission-band LIF technique (Lavieille et al., 2001).

Because long-lived phosphorescence is often quenched rapidly by O_2, Laser Induced Phosphorescence has not been used as commonly as LIF for flow diagnostics in liquids until recently. New findings by Nocera and his group (Ponce et al., 1993; Mortellaro and Nocera, 1996 ; Hartman et al., 1996) show that supramolecules may be designed to exhibit long-lived phosphorescence which is not quenched. A successful design by Ponce et al. (1993) indicates that the quenching of a lumophore (1-BrNp) can be prevented by mixing certain alcohols (indicated collectively by ROH) with an aqueous solution of glucosyl-β-cyclodextrin (Gβ-CD) that contains the lumophore. The resulting long-lived green phosphorescence has a typical lifetime of up to several milliseconds.

The availability of long-lived luminescent tracers has lead to the technique of Molecular Tagging Velocimetry (MTV) (Gendrich et al. 1997). As the molecular counterpart of Particle Image Velocimetry (PIV) technique, MTV utilizes molecules that can be turned into long-lived traces when excited by photons of appropriate wavelength. Typically, a pulsed laser is used to "tag" the regions of interest, and those regions are interrogated at two successive times within the lifetime of the tracer. The measured displacements of the tagged regions provide the estimates of fluid velocity vectors. By using the 1-BrNp·Gβ-CD·ROH complex as the long-lived tracer for MTV measurements in liquids, extensive studies have been conducted to investigate fundamental flow phenomena and applied fluid engineering problems over the past several years (Koochesfahani, 1999; Koochesfahani et al., 2000; Bohl et al., 2001; Lum et al., 2001).

Thomson and Maynes (2001) recently demonstrated the possibility of temperature measurement in a liquid by using Laser Induced Phosphorescence of the 1-BrNp·Gβ-CD·ROH complex. The method described by Thomson and Maynes (2001), however, is actually the phosphorescence version of the conventional single-dye LIF technique for temperature measurement, in that the emission intensity is used directly to infer the temperature. The spatial and temporal variations of the incident light intensity would have to be corrected for separately. These are the same issues that motivated the development of intensity-ratio-based LIF techniques.

In the present paper, a novel technique will be described for quantitative temperature mapping in a liquid by measuring the lifetime of laser induced phosphorescence. This methodology represents an extension of the MTV technique. As in MTV measurements, the phosphorescence

signal is imaged at two successive times within the lifetime of the tracer. The phosphorescence lifetime, estimated from the intensity ratio of the two images, is calibrated with respect to the temperature. This methodology also allows the simultaneous quantification of velocity and temperature fields in a fluid flow by using the same phosphorescent tracer (1-BrNp·Gβ-CD·ROH) and the same optical and equipment setup. While the displacements of the tagged regions between two interrogations provide the estimate of fluid velocity vectors, the intensity ratio of the two images represents the fluid temperature distribution. In the sections that follow, the details of the new technique are described along with a demonstration of its application to conduct simultaneous tempetaure and velocity measurement in a pulsed cold jet discharging into hot ambient fluid.

2. Description of the experimental technique

2.1 Technique basis

According to quantum theory (Pringsheim, 1949), the intensity decay of a photoluminescence process (either flourescence or phosphorescence) can be expressed in the form

$$I_{em} = I_o\, e^{-t/\tau}\,,\tag{1}$$

where the lifetime τ refers to the time when the intensity drops to 37% (i.e. $1/e$) of the initial intensity I_o.

For an excited state, the deactivation process may involve both radiative and non-radiactive pathways. The lifetime of the photoluminescence process, τ, is determined by the sum of all the deactivation rates:

$$\tau^{-1} = k_r + k_{nr}\,,\tag{2}$$

where k_r and k_{nr} are the radiative and non-radiative rate constants, respectively. According to photoluminescence kinetics, these rate constants are, in general, temperature-dependant (Ferreudi, 1987). The temperature dependence of the phosphorescence lifetime is the basis of the present technique.

The temperature dependance of the fluorescence lifetime of a probe has been used to measure the temperature of living cells (Chapman et al., 1995) and of liquid crystals (Huang et al., 1995). Since the fluorescence lifetime is always very short (on the order of nano seconds), the fluorescence lifetime is usually measured by oscilloscope-based instrumetation. Although Ni and Melton (1996) demonstrated the gase-phase temperature measurement using the fluorescence lifetime imaging technique, the experimental setup was relatively complicated when imaging the short fluorescence lifetime.

Brewster et al. (2001) have reported the possiblity of the temperature measurement in liquid by measuring the phosphorescence lifetime of a water-soluble phosphorscent dye, which is similar to the compound used here. The work of Brewster et al. (2001) is a single-point feasibility study using oscilloscope-based instrumetation. Futhermore, the compound utilized has a relatively short lifetime of 100 μs (at room temperature), nearly 50 times smaller than that reported herein, which makes it suitable for simultaneous velocimetry only for very high-speed water flows. The work described in the present paper represents, to our knowledge, the first planar temperature field measurements conducted in a liquid flow based on the direct imaging of phosphorescence lifetime with a conventional image detecting CCD camera.

2.2 Phosphorescence lifetime imaging

The total phosphorescence intensity I_p at a given point is given by

$$I_p = I_i\, C\, \varepsilon\, \Phi_p,\qquad(3)$$

where I_i is the local incident laser intensity, C is the concentration of phosphorescence dye, ε is the absorption coefficient, and Φ_p is the phosphorescence quantum efficiency.

The total phosphorescence intensity can also be separately determined from the integration of Equation (1) over all time, resulting in

$$I_p = \tau\, I_o \,.\qquad(4)$$

Now consider capturing the phosphorescence emission by a gated intensified CCD detector, where the integration starts at a delay time t_o after the laser excitation pulse with a gate period of δt. The phosphorescence signal S_p collected by the detector is then given by

$$S_p = \int_{t_o}^{t_o+\delta t} I_o\, e^{-t/\tau}\, dt \,.\qquad(5)$$

Using the equations given above, it can be shown that

$$S_p = I_i\, C\varepsilon\, \Phi_p \left(1 - e^{-\delta t/\tau}\right) e^{-t_o/\tau} \,.\qquad(6)$$

In the present study, the phosphorescence signal is imaged at two successive times within the lifetime of the tracer (Fig. 1). The first image is detected at the time $t = t_1$ after laser excitation for a gate period of δt to accumulate phosphorescence intensity S_1, while, the second image is detected at the time $t = t_1 + \Delta t$ for the same gate period to accumulate phosphorescence intensity S_2. The intensity ratio of the second image intensity (S_2) to the first image intensity (S_1) will be

$$\frac{S_2}{S_1} = e^{-\frac{\Delta t}{\tau}} \,.\qquad(7)$$

From Equation (7), it is evident that the intensity ratio of the two successive phosphorescence images is only a funtion of phosphorescence lifetime, and not the laser excitation intensity. This eliminates the effects of the variation of the incident illumination.

The phosphorescence lifetime can be calculated on a pixel-by-pixel basis from Equation (7) according to

$$\tau = \frac{\Delta t}{\ln(S_1 / S_2)},\qquad(8)$$

resulting in the distribution of the phosphorescence lifetime in a two-dimentional domain based on two sucessive phosphorescence images.
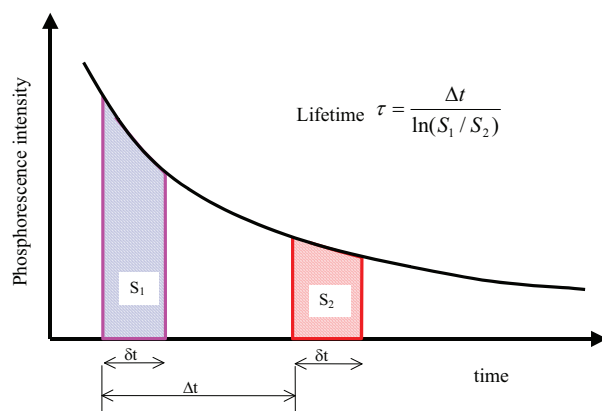

Fig. 1. Calculation of phosphorescence lifetime from two successive images.

2.3. Chemical composition

The phosphorescent tracer used in the present study is 1-BrNp·Gβ-CD·ROH complex, which has been used extensively as a long-lived tracer for MTV measurements. Further information about the chemical structure and photoluminescence performances of the 1-BrNp·Gβ-CD·ROH complex can be found from Ponce et al. (1993), Gendrich et al. (1997) and Koochesfahani (1999). Since the chemical composition of the complex affects the intensity and lifetime of the 1-BrNp·Gβ-CD·ROH complex significantly, we used a concentration of 2×10^{-4} M for Gβ-CD, a saturated (approximately 1×10^{-5} M) solution of 1-BrNp and a concentration of 0.06 M for the alcohol (ROH) in the present study, as suggested by Gendrich et al. (1997).

2.4. Experimental setup for calibration

Figure 2 shows the schematic setup for the calibration procedure to obtain the quantitative relationship between the temperature and the phosphorescence lifetime of the 1-BrNp·Gβ-CD·ROH complex. A Lambda-physik XeCl excimer ultraviolent (UV) laser (wavelength λ = 308 nm, energy 50 mJ/pulse, pulse width 20 ns) with appropriate optics was used to generate a laser sheet (thickness about 1 mm) to illuminate the cube-shaped test cell (about 3.0 liters in volume) containing an aqueous solution of 1-BrNp·Gβ-CD·ROH complex. Two faces of the cell were made of UV-transparent quartz. The apparatus was placed on a heating plate and a stirring rod was used to achieve thermal equilibrium in the test cell. A thermocouple (Hartscientific Model 1502A, temperature resolution ±0.01ºC) was placed in one corner of the apparatus to measure the actual temperature in the test cell. During the experiment, the temperature uniformity inside the test cell was checked and was found to be within 0.1ºC.

A 12-bit high-resolution (1280 × 1024 pixels) gated intensified CCD camera (PCO DICAM-Pro) with a fast decay phosphor (P46) was used in the present study to conduct phosphorescence image recording. The laser and the camera were synchronized using a digital delay generator (SRS DDG535), which controlled the timing of the laser sheet illumination and the CCD camera data acquisition. The phosphorescence images captured by the CCD camera were subsequently transferred to a host computer for analysis.
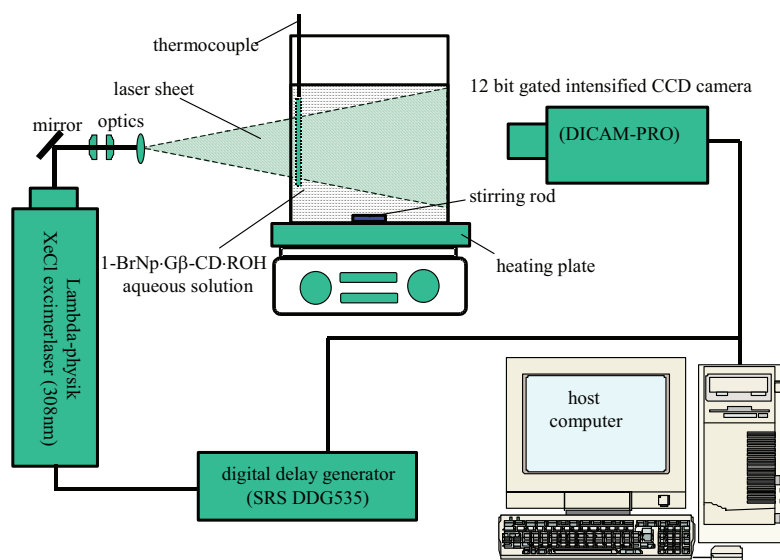

Fig. 2. Schematic setup for the calibration procedure.

2.5. Phosphorescence lifetime of the 1-BrNp·Gβ-CD·ROH complex as a function of temperature.

To acquire temperature data, the aqueous solution of the 1-BrNp·Gβ-CD·ROH complex was first heated to a predetermined temperature level. After thermal equilibrium was established, the phosphorescence intensity decay profile was measured by changing the time delay of the camera image acquisition after the laser excitation with a fixed gate period ($\delta t = 500 \mu s$ for the present study). Figure 3(a) shows the decay curves (in log-linear scale) of the phosphorescence intensity of 1-BrNp·Gβ-CD·ROH complex in the temperature range of 22.5ºC – 50ºC. It is found that all the phosphorescence decay curves at different temperature levels are almost single-exponential curves, which is typical of phosphorescence decay behavior, as expected theoretically. Therefore, Equation (8) can be used to calculate the phosphorescence lifetime of the 1-BrNp·Gβ-CD·ROH complex.

Based on the intensity decay curves, the phosphorescence lifetimes of the 1-BrNp·Gβ-CD·ROH complex at different temperatures were calculated. The results are shown in Fig. 3(b). It can be seen that the lifetime varies significantly with increasing temperature, dropping from about 4.6 ms to 0.4 ms as the temperature changes from 20.0ºC to 50.0ºC. The relative temperature sensitivity of the phosphorescence lifetime ranges 5.0% per degree at 20.0ºC to 20.0% per degree at 50.0ºC. The temperature sensitivity is much higher than those of most commonly used LIF dyes (for example, the temperature sensitivity of Rhodamine B is about 2.0% per degree).

2.6. Simultaneous temperature and velocity measurements

One of the advantages of the present technique is that it allows the simultaneous quantification of velocity and temperature fields in fluid flows by using the same phosphorescent tracer (1-BrNp·Gβ-CD·ROH) and the same optical and equipment setup. As described above, the phosphorescence signal is imaged at two successive times within the lifetime of the tracer. The phosphorescence lifetime, estimated from the intensity ratio of the two successive images by using Equation (8), can be used to get the temperature distribution by using the relationship between the phosphorescence lifetime and temperature. Meanwhile, the fluid velocity vectors can be estimated from the displacements of the tagged regions between two interrogations through a standard MTV image processing routine (Gendrich and Koochesfahani, 1996 ; Koochesfahani, 1999).
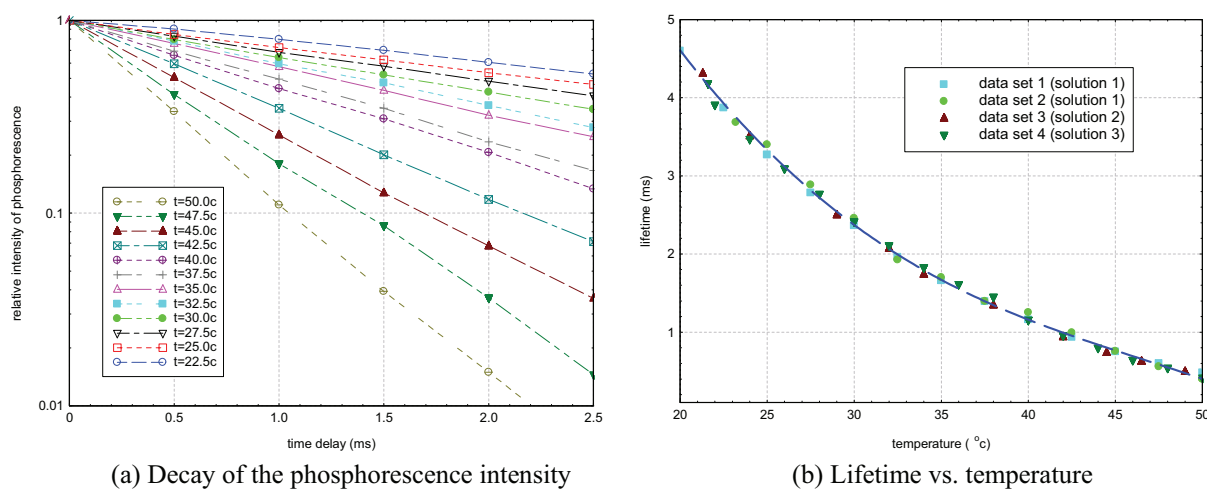

(a) Decay of the phosphorescence intensity   (b) Lifetime vs. temperature

Fig. 3. Calibration results.

3. Demonstration experiment

In order to demonstrate the feasibility of the technique described above, we set up a demonstration experiment to conduct simultaneous temperature and velocity measurements in a pulsed jet flow. Figure 4 shows a schematic setup of the demonstration experiment, where a cold pulsed jet exhausts into hot ambient fluid. A turkey baster is used in the present study to generate the cold pulsed jet flow. The diameter of the turkey baster at the exit is about 6.0 mm (D=6.0 mm). The temperature of the 1-BrNp·Gβ-CD·ROH aqueous solution in the turkey baster is about 25.0ºC, while the temperature of the 1-BrNp·Gβ-CD·ROH aqueous solution in the container is about 36.0ºC. The cold fluid in the

Fig. 4. Schematic setup of the demonstration experiment.

turkey baster exhausts into the hot ambient fluid as a pulsed jet by manually squeezing the rubber bulb at the end of the turkey baster. The maximum jet velocity at the exit of the nozzle is about 0.27 m/s. The test section is illuminated with a XeCl excimer UV laser sheet (wavelength λ = 308 nm, energy 50 mJ/pulse, pulse width 20 ns). A beam blocker is used to turn the UV laser sheet into multiple laser beams to tag the fluid.
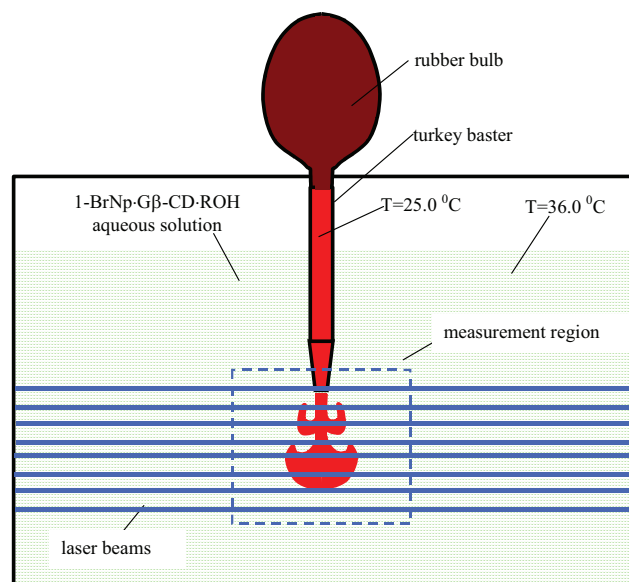
The 12-bit high-resolution intensified CCD camera (PCO DiCAM-Pro) is used to conduct the phosphorescence image recording. The camera is operated in the dual-frame mode, where two full-frame images of phosphorescence are acquired in quick succession from the same laser excitation pulse. For the present demonstration experiment, the first image is captured at the time delay of 500 µs after the laser pulse with the interrogation gate period of 500 µs. The second phosphorescence image is captured at the time delay of 3.5 ms after the laser excitation with the same interrogation gate period. A typical pair of first and second phosphorscence images is given in Fig. 5.

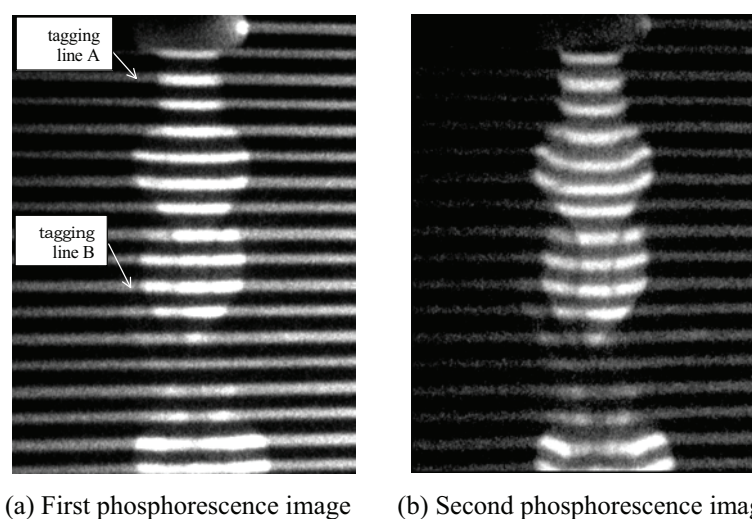(a) First phosphorescence image   (b) Second phosphorescence image

Fig. 5. Typical pair of first and second phosphorscence images.

From the comparison of the first and second phosphorescence images, it can be seen that the tagging lines in the first image have deformed into curved lines in the second image. The velocity distribution of the flow field can be obtained from the displacements of the corresponding lines between the first image and second image through a standard line-tagging MTV image processing routine (Koochesfahani, 1999). Since the temperature of ambient fluid (T=36.0ºC) is higher than the pure jet flow (T=25.0ºC), the phosphorescence intensity of the hot ambient fluid decays faster than that in the cold pure jet. Therefore, the portions of the tagged lines in the hot ambient fluid are much dimmer than that in the cold pure jet in the second image. The phosphorescence lifetime distributions can be obtained from the phosphorescence intensity ratios of the first and second phosphorescence images based on Equation (8). By using the relationship between the temperature and phosphorescence lifetime of 1-BrNp·Gβ-CD·ROH complex, the temperature distribution can be constructed.

For the present line-tagging MTV experiment, the averaged phosphorescence intensities of 15×15 pixels windows at the center of the tagging lines are used for the lifetime calculation. In order to match to the corresponding pixels in the first and second phosphorescence images, the jet flow is assumed to be one-dimensional. However, the jet flow is not one-dimensional in reality, the mismatch of corresponding pixels may result in measurement error. Further discussion about the measurement error due to the mismatch is given in section 4.3.

Sample simultaneous temperature and velocity data are shown in Fig. 6 in terms of quantitative transverse profiles along tagging lines A and B, marked in the Fig. 5(a). Along tagging line A, which is located about 0.4 diameters downstream from the nozzle exit, the velocity and temperature distributions were found to have nearly top-hat profiles, as expected. At the further downstream location of Y/D=3.5 (tagging line B), some hot ambient fluid has been engulfed into the pure jet through the entrainment process between the pure jet and ambient fluid. The top-hat velocity profile is rounded up and the velocity at the center of the jet decreases from 0.27 m/s at the nozzle exit to 0.20 m/s. The temperature of the pure jet flow also increases to about 27ºC at this downstream location. There are two temperature peaks in the transverse temperature profile of the pure jet, which is due to the engulfing of hot ambient fluid from the two sides of the pure jet flow. The engulfing of the hot ambient fluids to form two temperature peaks in the pure jet is visualized clearly in the phosphorescence images shown in Fig. 5, which are the two dark regions in the pure jet flow.
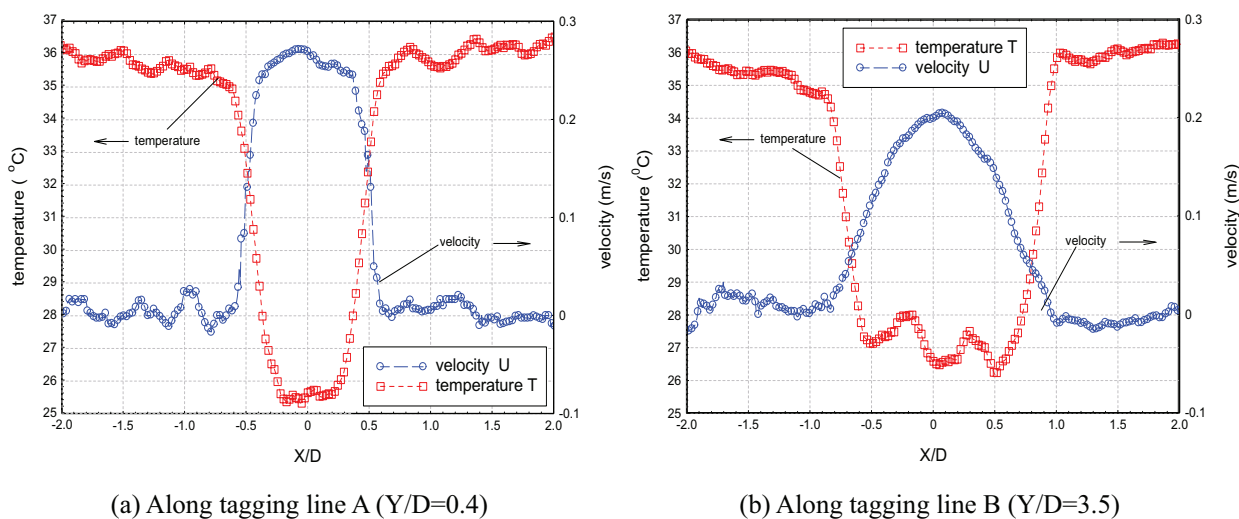

(a) Along tagging line A (Y/D=0.4)   (b) Along tagging line B (Y/D=3.5)

Fig. 6. Simultanous velocity and temperature profiles along tagging lines A and B.

4. Temperature measurement accuracy

Several factors may affect the accuracy of the temperature measurement when using the present method. These factors include:

4.1. Noise in the phosphorescence image

The noise in the instantaneous phosphorescence images may result in the measurement error of temperature when using the present method. This error varies with the sensitivity and available bits of the camera. For the 12-bit high resolution camera (DICAM-PRO) used in the present study, the standard deviation of the phosphorescence lifetime σ_τ due to the noise in the phosphorescence image is within 1.0%, as estimated by the equation $\sigma_\tau = -\dfrac{\Delta t}{(\ln(S_1/S_2))^2}\sqrt{(\dfrac{\sigma_{S_1}}{S_1})^2 + (\dfrac{\sigma_{S_2}}{S_2})^2}$ suggested by Ballew and Demas (1999), where σ_{S_1} and σ_{S_2} are the standard deviations of S_1 and S_2, respectively. Therefore, the temperature error due to the image noise is estimated to be less than 0.25ºC in the range of 20.0ºC to 50.0ºC.

4.2. Single exponential model of the phosphorescence intensity decay

As shown in Fig. 3, the decay profile of the phosphorescence intensity may have small deviation from the single exponential curve. Therefore, the phosphorescence lifetime value estimated from the best fit to a single exponential decay profile might be slightly different from the lifetime value as estimated from the two phosphorescence images by using equation (8). Using the lifetime-temperature calibration curves from the best fit to interpret the phosphorescence lifetime map calculated from the intensity ratio of two phosphorescence images may result in measurement error. This error can be as big as 0.25ºC in the temperature range of 20.0ºC to 50.0ºC.

Rather than using a best fit calibration procedure, a two-point lifetime-temperature calibration procedure, which is used in the present demonstration experiment, can solve this problem. The two-point calibration procedure, uses the parameter settings of the phosphorescence image acquisition for the lifetime-temperature calibration and the real experiment, and therefore can eliminate this error completely.

4.3. Mismatch of the corresponding pixels in the first and second phosphorescence images

During the time interval between the two phosphorescence images (usually on the order of several milli-seconds), the corresponding pixels in the first and second phosphorescence images moves several pixels. The mismatch of the corresponding pixels can result in measurement error of temperature field. For the line-type tagging experiment shown in Fig. 6, this error is estimated to be as big as 0.25ºC in the shear layer regions. Using a grid-type tagging method to solve the ambiguity of the corresponding pixels in the two phosphorescence images can minimize this error.

5. Conclusions

A novel method for conducting quantitative temperature field measurements in liquid has been developed by taking advantage of the phosphorescence lifetime dependence on temperature. 1-BrNp·Gβ-CD·ROH complex was used as the long-lived phosphorescent tracer in the present study. The decay rate of the phosphorescence intensity, in terms of the phosphorescence lifetime, was found to change significantly with the temperature.

The methodology described in the present study represents an extension of the MTV technique. As in MTV measurements, the phosphorescence signal is imaged at two successive times within the lifetime of the tracer. The phosphorescence lifetime, estimated from the intensity ratio of the two images at each point, is calibrated with respect to temperature. This methodology allows the simultaneous quantification of velocity and temperature fields in a fluid flow by using the same phosphorescent and same optical and equipment setup. While the displacements of the tagged regions between two interrogations provide the estimate of fluid velocity vectors, the intensity ratio of the two images represents the fluid temperature distribution. The implementation and application of this technique are demonstrated here by the simultaneous temperature and velocity measurements in a pulsed cold jet discharging into hot ambient fluid.

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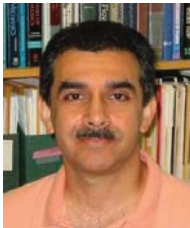
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