An analysis of long term temperature measurement using laser induced fluorescence

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Abstract. The temperature measurement is extremely important because it occurs in many technical and engineering processes, including combustion chambers, mixers or chemical reactors as well as environmental flows. In contrast to the point measurement method, Laser Induced Fluorescence (LIF) allows temperature determination in the whole plain 2D, or even 3D, domain. A major advantage of LIF is also its relatively high accuracy. This technique involves dissolving a temperature-sensitive fluorescence dye to a fluid. It is known that in LIF the fluorescent re-emission is a function of temperature, but in many cases, it can also be a function of time, due to dye properties degradation. In the present research, a long-term temperature measurement using LIF was performed in order to analyse the method uncertainty related to time. The results of the stability of Rhodamine-B in non-isothermal experimental measurements in water solution, together with the chemical analysis using spectrophotometry, are presented.

1. Introduction

In many engineering applications acquiring specific information about the behaviour of a process, it is possible to developed only by measuring the scalar quantities (temperature, concentrations). Such information is not only important in macro-scale, but also in micro-scale [1]. Devices such as combustion chambers, heat exchangers, cooling/heating systems, mixers or chemical reactors are of most interest. The temperature knowledge allows to design or redesign them in a responsible way. Because of the fact that one of the most essential issues in thermo-fluid engineering is heat transfer, the temperature measurement, being even of greater importance than the velocity field, and for this reason is widely researched [2-4].

There are many techniques that could be used in order to obtain the spatial distribution of the temperature in fluids [5]. One of the most popular ones is the thermocouple (TC). An appropriate calibration of the TC probe provides the measuring temperature precision at 0.1 K. However, there are some limitations of the use of TC probes in temperature measurements. A major drawback is the fact that a TC is able to measure the temperature only at a single point. Under this circumstance, determining the temperature field in the analysed fluid would require a large number of sensors placed within. For this reason, some “non-intrusive” methods were developed in the past. One of the full field measurement methods uses encapsulated thermo-chromic liquid crystal particles (TLC) as a probe. The TLC technique takes advantage of the dependence between temperature and their optical properties. TLC is illuminated and then reflects light at a visible wavelength depending on the local

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temperature. The accuracy of the measurement with TLC depends largely on the calibration process. The accuracy relative to the 6°C temperature range is approximately 0.6°C [2]. Encapsulated liquid crystal particles can be used to measure temperature inside fluids. The major drawback of this technique is a narrow temperature range, strong nonlinearity and a significant time response of the relatively large particles.

One of the alternatives for TLC is laser induced fluorescence (LIF) [6]. This technique involves dissolving a temperature-sensitive fluorescence dye to a fluid (gas or liquid) and then illuminating the fluorescent dye by laser. Fluorescent dyes have a property to absorb light energy and generate re-emission at different frequencies [7]. LIF assumes that the re-emission is a function of temperature (typically linear). In the case of strong intensity of the primary light and a sensitive camera, it is possible to obtain a temperature field of the fluid at once. A major advantage of LIF is a large temperature range (limited by the used dye type) and better accuracy in comparison to TLC, i.e. typically about 1.5 °C for a 40°C temperature range. LIF can be also used with the PIV method [7-11].

Depending on the nature of the fluid to be analysed and the type of measurement, there are various types of dyes that could be used for the LIF technique. One of the most widely used fluorescent dyes for measuring temperature and concentration in liquids is Rhodamine-B. It is characterized by high solubility in water, high absorption, emission, and high sensitivities when excited with Nd:YAG laser (532 nm). The high power output of the device allows it to work freely within the whole measured area and its very short pulse duration reduces noise at precisely captured images [5-7].

The temperature range and gradients are of the most importance among the measuring methods, yet accuracy cannot be omitted. LIF, as a “non-intrusive” temperature mapping technique, ensures acceptable temperature readings in relation to the temperature range. One of the natural features of soluble organic dyes used in this method is a measurable decrease in the quantum yield, caused by an increase of the quenching effect, proportionally to the solution’s temperature increase. Moreover, the size of the dye molecules (about 10 nm) is what makes them perfect even for microscale measurements [1]. The LIF method also has disadvantages. The results need light and in general all optical conditions to remain constant, which is difficult to obtain. A lot of imperfection follows the image recording [12-14]. Images are not free from the possibility of bias caused by the background noises [4]. These are problems connected with the laser, whose light intensity might vary in time. Additionally, the error of the shadowgraph effect can occur near the non-isothermal walls [15-17].

The LIF method requires that the dye concentration together with the light intensity be constant because they are directly reflected in the temperature measurement. However, even the dye concentration is constant the dye re-emission (which a function of the temperature) can be function of time due to dye properties degradation. This behaviour will be analysed in the present paper in long-term temperature measurements. The influence of this effect on the temperature measurement will be verified in order to obtain the method uncertainty.

2. Experimental set-up and data evaluation
Temperature measurements were performed in two cavities presented in Figure 1. The first, rectangular cavity was a part of an open system, while the second, cubical cavity was a classical benchmark cavity for natural convection with heated and cooled vertical walls.

![Figure 1. The geometry of measurement section (a) opened cavity (b) closed cavity.](image-url)
In order to obtain the LIF signal, a fluorescent dye was exposed to laser light at a specified wavelength. The ration of the energy emitted by the dye molecules per absorbed energy is called quantum efficiency \( \Phi \) and the total emitted fluorescent energy can be defined as:

\[
I = \varepsilon A I_o C \Phi \frac{\lambda_c}{\lambda_f}
\]

where \( I_o \) is the incident light flux, \( C \) is the concentration of the fluorescent dye (in the present paper with solution 1000, 800 and 500 \( \mu \)g/dm\(^3\)), \( \varepsilon \) is the molar absorptivity, \( A \) is the fraction of the collected available light. The wavelength ratio \( \lambda_c/\lambda_f \) accounts for the energy loss in the excited state. The quantum \( \Phi \) may result in temperature dependent dye intensity. The observed temperature effect on the quantum yield is possibly induced by quenching effects. With the increasing temperature the quenching effect increases. In the present case, the LIF signal for Rhodamine B decreases by about 2% for 1K decrease. In order to obtain a signal to temperature dependency other parameters, such as dye concentration, light intensity and camera settings have to be constant. A schematic diagram of an experimental set-up has been shown in Figure 2. In the performed measurements a charge coupled device (CCD) camera with a long pass at 540 nm was arranged perpendicular to the Nd:YAG laser light sheet (\( \lambda_e=532 \) nm with pulse energy of 40 mJ). This arrangement is most preferred as it ensures the lowest optical distortion.

![Figure 2. Schematic diagram of the experimental set-up.](image)

In the case of the open rectangular cavity (75x75x30mm) measurement, a water bath thermostat was used in order to keep the temperature in the enclosure at the specified level. For the cubical closed enclosure of 32x32x32mm with a heated vertical wall, opposite a cooled one as well as an adiabatic walls, in order to keep the cold wall at a constant temperature, water bath thermostat was used, while for the heated wall, a resistance heater was used. An accurate temperature measurement was performed with thermocouples located at the walls (closed enclosure) and with thermocouples mounted at the inlet and outlet of the open cavity. The temperature range considered here varied from 23,5°C up to 50°C.

![Figure 3. Data evaluation procedure for LIF measurement.](image)
The data evaluation procedure, consisting of 6 steps, is presented in Figure 3. After recording a series of raw images (typically up to 100 images for statistical analysis) and averaging them, the background image (dark image) was subtracted from the raw images in order to obtain the LIF signal.

Almost all the light sources generate inhomogeneities in the formed light sheet. This effect is corrected in the next step, i.e. sheet correction. The recorded information is normalised with the sheet images (images recorded at uniform temperature) and the result is a highly uniform output signal. Because the normalised images may contain local noise, due to fluid or glass impurities, it is common to use a digital filter to correct and smooth any local impurities. In the last step, the local intensity was mapped into absolute temperature. To do that, the calibration curve had to be created based on the measurements at a known, uniform and constant temperature. Calibration images, as typical LIF images, include not only the LIF signal, but also any distortion in the light, optics, signal detection as well as physical distortion due to fluid density variation, self-absorption and shadowgraph effects.

Some sources of errors are eliminated with statistical operations, for example pulse-to-pulse laser light variance, while some other effects, however not all, can be eliminated through employing noise filters.

3. Experimental results

In the current experiments Rhodamine 610 chloride, (Rhodamine B), (CAS No.: 81-88-9) was used in order to perform single colour the LIF and the chemical spectroscopy analysis. Chemical structure of the Rhodamine B (610) used in the experiment is presented in Figure 4. Deionised water (<0.07 S/cm), used to prepare standard and sample solutions was obtained from the HLP5 pure water system (Hydrolab, Gdansk, Poland). Pure methanol was used for cleaning of the glass.

The detailed concentration of analysed rhodamine was determined using UV-Visible Spectrometer (Cintra 10, GBC Scientific Equipment). All experiments were conducted in aqueous solution. The measurements of absorbance of rhodamine solutions were conducted at wavelength 553.5 nm from which the concentrations were calculated. The fixed wavelength for measurements was selected for maximum of absorbance achieved in the wavelength scanning of rhodamine solution 1000 μg/l in the range from 370 nm to 700 nm, at scan speed 1.000 nm/min and data interval of 1.04 nm.

Figure 4. Chemical structures of Rhodamine 610 (left) and absorption spectrum of rhodamine solution in the range 370 – 700 nm (right).

The absorption spectrum is presented in Figure 4 (right). Calibration was performed against rhodamine standard diluted in water in the range 300–1000 μg/l. The value of correlation coefficient (R²) was 0.9952 and confirmed a good linearity of the method. The intra-day accuracy of the method determined by calculating the agreement between the measured and known concentration (reference concentration 1000 μg/l) was 98.4 %. The intra-day precision of the method determined by calculating the relative standard deviation (RSD) of the replicated measurements (n = 4) was 1.7 %. The inter-day precision of the method calculated using quality control (QC) samples at concentrations of 1000 μg/l (n = 5) was 1.9 %.
Figure 5 shows sets of four post-process images according to the data evaluation procedure presented before, for laser induced fluorescence. The first picture 5(a) shows a raw image, while the next images were obtained after background subtraction 5(b), normalisation 5(c) and correction 5(d), according to the evaluation procedure presented in Figure 4. As a final result, uniform intensity for a uniform temperature was observed. A series of the presented analyses using experimental measurements was conducted for different temperatures and time period (up to 3 months).

![Figure 5](attachment:image.png)

Figure 5. Experimental images: raw image (a), image after background subtraction (b), normalised image (c) and corrected image (d).

The correlation between the fluorescent intensity and temperature is necessary in order to use this method for temperature measurements. To obtain this correlation, a series of measurements was performed for a known temperature. The results of the calibration are presented in Figure 6(a) showing a monotonic intensity decrease with the increasing temperature. In order to ensure repeatability, the calibration was repeated for the same temperature range and for different sub-regions and spatial resolutions. In order to perform spectrophotometric analysis, the calibration curve of the absorption presented in Figure 6(b) was created against rhodamine diluted in water in the range 300–1000 µg. In Figure 6(c) and 6(d), the intensity signal for the whole analysed region at uniform temperature 25.0 and 50.0°C versus time has been presented. It can be seen that, for the open cavity, during the first months, the intensity signal from Rhodamine-B increases. This can be caused by chemical and photochemical decomposition as well as by impurities (open system). For a longer time period, significant intensity decreases with time was observed. The intensity decrease in reference to the primary intensity is significant and, at the end of the experiment, the signal from the fluorescent dye is 15-20% lower. The presented measurements reveal experimental uncertainties related to dye properties degradation in time of 25-30% for temperature measurement.
In the Figure 7 the absorbance of Rhodamine-B solution 1000/800 µg/l at 553.5 nm versus time is presented. In this measurements almost uniform absorption was observed for a first five days, but for longer time period the absorbance significantly decrease. At the end of measurement series after 7 weeks decrease in absorbance was about 23% in reference to primary absorbance. It is interesting to notice that in both presented analyses in open cavity and in spectrophotometric analysis Rhodamine-B properties significant deprecate.

![Figure 6](image_url)

**Figure 6.** Calibration curve for LIF (a) and spectrophotometry (b) and intensity signal at uniform temperature 25°C (c) and 50°C (d).

![Figure 7](image_url)

**Figure 7.** The absorbance of rhodamine solution at 553.5 nm versus time.
The difference in results for the first period can be caused by several reasons, i.e., longer dye exposition in cavity, natural impurities, water evaporation, bubbles generation in open system. In the open system, total two litters of rhodamine solution was in circulation system (including, heaters, pumps).

![Intensity vs Temperature](image1)

![Intensity vs Time](image2)

**Figure 8.** Rhodamine solution intensity at different temperature (left) and rhodamine intensity versus time at uniform temperature 23.5°C (right).

In order to avoid all problems concerning opened system, a new system with closed cavity (containing about 30 ml of rhodamine solution) was constructed. In Figure 8 (left) calibration curve base on the results from different days it is shown. In Figure 8 (right) results for first 3 weeks and for closed cavity are presented. It can be seen in Figure 8 that during this period intensity signal increases independent of images sub-region or filtering method.

4. Conclusion

The use of LIF for a non-invasive temperature measurement has become a widely used technique. Various correction procedures based on the digital imaging technology have been proposed in order to decrease the measurement error. In the present paper, the temperature measurement with the use of the LIF method was investigated in order to evaluate the possible dye properties degradation in a long-term temperature measurement using LIF. The results of the stability of Rhodamine in non-isothermal in a closed and open cavity together with a chemical analysis using spectrophotometry were presented. The data evaluation procedure with distortion elimination processes were presented for the measurement in the open and closed cavity system. The experimental uncertainties associated with the dye chemical variation were evaluated. Due to intensity variations between different experiments, the intensity fluctuations unrelated to dye properties degradation were observed. This effect occurs in the real experimental measurements with cavities as well as in the chemical analysis. It was noticed that after 2-3 months, the intensity signal from the fluorescent dye first slightly increases and then, after about 2 weeks, begins a constant decrease.

The intensity decrease in reference to the primary intensity was significant and, at the end of the experiment, the signal from the fluorescent dye was 15-20% lower. The presented measurements reveal experimental uncertainties related to dye properties degradation in time of 25-30% for temperature measurement. Because the fluorescence intensity is not only dependent on temperature, several normalisation procedures are used. For a long-term analysis, an extra correction step related to intensity variation in time has to be made.

These effects, unlike others, cannot be minimized by averaging or any statistical analysis. To avoid possible additional correction, a series of short-term analyses should be performed. Two-colour Laser Induced Fluorescence eliminates the major error sources of laser-light fluctuations in the flow field.
because the signal is proportional to the ratio between the temperature-sensitive and the temperature insensitive fluorescent dyes and the dye degradation effect related to time may not been observed. Such situation may occur only if the two different dyes change their properties with time in a similar manner.

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