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TEMPERATURE DEPENDENCE OF GAS-PHASE ACETONE PHOTOLUMINESCENCE UNDER LOW PRESSURE CONDITIONS AT SEVERAL EXCITATION WAVELENGTHS

Slaven Bajić¹, Marcos Rojas-Cárdenas¹, Christine Barrot Lattes¹, Stéphane Colin¹

¹ Institut Clément Ader (ICA), Université de Toulouse CNRS, INSA, ISAE-SUPAERO, Mines-Albi,
UPS, Toulouse, France

slaven.bajic@univ-tlse3.fr, rojascar@insa-toulouse.fr, lattes@insa-toulouse.fr, colin@insa-toulouse.fr

KEY WORDS

molecular tagging thermometry (MTT), laser induced fluorescence (LIF), absorptiometry, rarefied gases, microfluidics

ABSTRACT

1. Introduction

With the accelerated development of microfabrication techniques and micro-electro-mechanical systems (MEMS) during the past few decades, gas microfluidic devices have gained an important role in various research domains, such as aeronautics [1], space [2], medicine [3] and electronics [4].

In these devices, the flow dynamics are strongly impacted by the effect of gas rarefaction, whose level is defined by the Knudsen number $Kn = \lambda / L_c$, where λ represents the molecular mean free path and L_c the characteristic dimension of the system. Many of the gas microfluidic devices rely on the unique flow behavior that occurs in the moderately rarefied regime, also known as slip-flow regime, for which Kn ranges from 10^{-3} to 10^{-1} . In this regime, two main effects occur at the channel walls and govern the flow behavior: velocity slip and temperature jump [5]. Both phenomena make the classical continuum theory not adapted to describe precisely the flow, leading to the necessity to introduce new boundary conditions in the theoretical models. To validate these models, there is a significant lack of experimental research due to the difficulties in obtaining temperature and velocity profile measurements within a microsystem in a non-intrusive manner. This presents a major impact on speed and efficiency of the development of gas microfluidic devices operating in the slip-flow regime, as the prototype development and optimization process relies on theoretical models.

Molecular tagging technique has been identified as a suitable candidate for low-intrusive temperature and velocity profile measurements in a microchannel gas flow. The technique is based on the photoluminescence phenomenon: the gas is seeded by the tracer molecules which emit light signal in form of fluorescence or phosphorescence upon laser-light excitation. The emitted light signal intensity and its temporal evolution are then extracted in order to determine physical properties of the gas, such as temperature and/or velocity.

This study will focus on the molecular tagging for temperature measurements, also referred to as molecular tagging thermometry (MTT). Due to its low toxicity, high vapor pressure and its vast use in laser-induced fluorescence (LIF) [6], acetone has been chosen as the most suitable tracer molecule for this study. Even though the photoluminescent properties of acetone have been well investigated, there

is still a deficit of research performed on acetone photoluminescence at low pressures and low gas concentrations, which would correspond to a rarefied gas regime. Therefore, before applying the molecular tagging to the measurement of the temperature profile in a microchannel gas flow, it is necessary to evaluate the photoluminescent properties of acetone at low pressures.

In that regard, the main goal of the present study is to estimate the temperature dependence of acetone photoluminescent properties, such as fluorescence light intensity, absorption cross-section and fluorescence quantum yield, under low-pressure conditions at various excitation wavelengths and 3 different emission wavelength ranges, in the narrow temperature range from 20 to 100°C, using the molecular tagging technique. Ultimately, the results should serve as a vital tool in the development of the optimal experimental strategy for the temperature measurements in gas microfluidic devices and for the validation of the existing theoretical models.

2. Experimental setup

An experimental setup for the photoluminescence intensity and light absorption measurement of gas mixtures under various operating conditions has been designed. The setup incorporates 3 main sub-systems: static test cell, optical system and gas control system, as presented in Fig. 1. The core of the experimental setup, the aluminum static test cell, consists of cross-shaped, 55 mm long and 8.6 mm diameter cylindrical cavities which are sealed by ISO-KF Suprasil[®] windows from the sides and by an ISO-KF borosilicate glass window on the top. On the bottom side, the cell is connected to the gas control system using ISO-KF vacuum flanges. In the bottom cavity, 2 different temperature sensors, type K thermocouple and NTC thermistor are positioned near the laser impact zone. For the pressure monitoring during the experiments, 2 capacitive gauge pressure sensors with different sensitivities are incorporated in the gas control system near the test cell. Heating of the gas in the test cell is achieved using the resistance heaters attached to the external sides of the test cell.

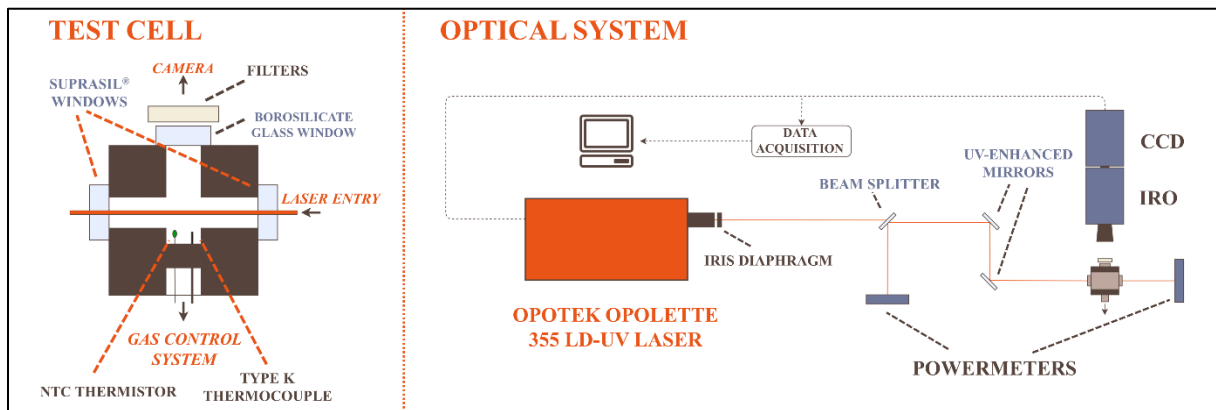


Figure 1: Schematic of the experimental setup for the study of acetone fluorescence.

For the excitation of the gas mixture within the test cell, an *Opotek Opolette*[®] 355LD-UV variable excitation wavelength laser is used. The emitted pulsed laser beam is guided towards the test cell using a set of optical elements and its energy is measured using two *Gentec*[®] QE12LP-S-MB joulemeters before and after the test cell, enabling the calculation of the gas light absorption. Photoluminescence image is captured by the image acquisition system comprising 105 mm *Nikkor*[®] lens, an intensified relay optics (IRO) and an *Imager Intense* CCD camera by *LaVision*[®]. Laser pulses are synchronized with the camera trigger by means of the programmable timing unit (PTU). With the aim of estimating the temperature sensitivity of the photoluminescence in different emission ranges, a combination of interposed 450 nm short-pass and 435 nm long-pass filters was placed on top of the test cell.

Gas mixture content in the test cell was controlled using a modular gas control system assembled using ISO-KF vacuum element. Prior to the experiments, the test cell and the gas control system have been

tested for any eventual leaks. The hourly leak rate was found to have insignificant impact on the molecular concentration in the test cell (< 0.5 % at 3000 Pa). As the main sources of the measurement uncertainty at this low pressure, camera read noise, adsorption / outgassing of acetone vapor by the cell walls and acetone photolysis were identified.

The images of the pure acetone fluorescence were acquired at 5 excitation wavelengths from 275 nm to 320 nm, at a delay time of 5 ns after the laser pulse and with a 100 ns gate time. With the purpose of increasing the signal-to-noise ratio (SNR), 150 images were integrated in an individual image using the on-chip integration technique. Subsequently, 20 of the individual images were averaged to obtain the final image. A list of the experimental conditions that have been investigated in this work is given in Tab. 1.

Laser excitation wavelength [nm]	275	289	300	310	320
Gas pressure [Pa]	3000 ± 15				
Gas Temperature [°C]	22 ± 0.5		45 ± 0.5		85 ± 1

Table 1: List of the investigated experimental conditions for the study of pure acetone fluorescence.

3. Results and conclusions

Equation (1) results from the widely accepted model of the acetone fluorescence [7] applied to the present study, where S_{exp} (photons) is the recorded mean intensity of the region of interest (ROI), E_{exp} is the laser beam energy entering the test cell (J), λ is the laser excitation wavelength (m), hc is the energy of a photon (J m), P is the gas pressure (Pa), T is the gas temperature (K), k is the Boltzmann constant (J / K) and η_{opt} is the efficiency of the collection optics (-). S^* (photons / (Pa / J)) designates the quantity proportionate to the captured fluorescent signal intensity per molecule per number of sent photons, a measurement of the molecular potential to fluoresce at a particular set of conditions, such as laser excitation wavelength, gas pressure and gas temperature. The absorption cross section σ (m²) is the probability that a photon passing through the molecule will be absorbed by that molecule multiplied by the average cross-sectional area, while the filtered fluorescence quantum yield Φ_{filt} (-) is the probability of the excited molecule to relax in the form of fluorescence in the observed emission range.

$$S^*(\lambda, T, P) = \frac{S_{exp}}{\left(\frac{E_{exp} \lambda}{h c}\right) \left(\frac{P}{k T}\right)} \propto \eta_{opt}(\lambda_{emission}) \sigma(\lambda, T) \Phi_{filt}(\lambda, P, T) \quad (1)$$

In accordance with the presented model, captured fluorescence images were normalized by the quantities $((E_{exp} \lambda) / (h c))$ and $(P / (k T))$ in order to obtain a comparable image. Moreover, background images were subtracted and ROI's were identified. As a result, a final image as shown in Fig. 2a was obtained. Mean value of the ROI's from the final image was used to evaluate and compare the acetone fluorescence intensity amongst different conditions. Fig. 2b shows the variation of the acetone fluorescence with the laser excitation wavelength for 3 temperatures in the 3 emission regions. Overall, the fluorescence intensity decreases with the increase of gas temperature in all emission ranges, with the effect becoming stronger for the lower excitation wavelengths. An interesting observation from these results is the fact that the sensitivity of the acetone fluorescence to temperature increase is more significant in the narrow emission range from 435 nm to 450 nm. This means that the observation of the acetone luminescence in the specific emission bandwidths and at lower excitation wavelengths could increase the accuracy of the gas thermometry using molecular tagging technique.

In order to validate the observations from these results, experiments at additional pressures and temperatures will be conducted. Furthermore, additional improvements to the experimental procedure will be implemented in order to increase the precision and reduce the uncertainty of measurements. Nevertheless, the observed results are encouraging, as they indicate that with some setup improvements and appropriate temperature measurement strategies (dual-wavelength, two-color), high precision thermometry in rarefied gas flows could be achieved.

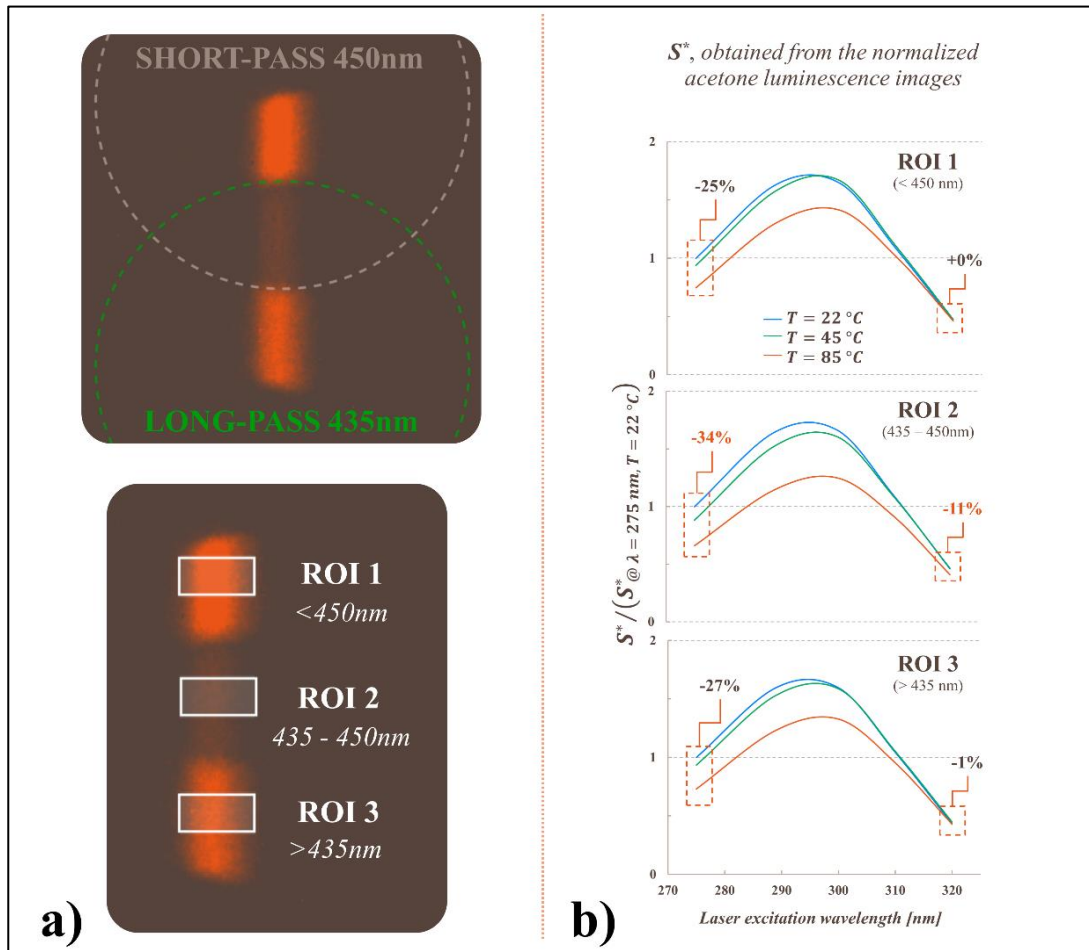


Figure 2: a) Sample processed images of the filtered acetone fluorescence S^* in the 3 spectral regions of interest (ROI). b) Variation of the filtered acetone fluorescence S^* normalized to the $T=22^\circ\text{C}$ and $\lambda=275\text{nm}$, in respect to the laser excitation wavelength and for the 3 spectral ROI's.

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