Velocity field measurements in gas phase internal flows by molecular tagging velocimetry

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Abstract. The goal of the present work is to implement Molecular Tagging Velocimetry (MTV) for the analysis of internal gas flows in mini-channels. A MTV experimental setup has been designed. Tagging and detecting steps are respectively insured by a UV laser and a CCD camera coupled to intensified relay optics. A specific channel with 1 × 5 mm\(^2\) rectangular cross section has been designed and equipped with integrated temperature sensors along its 20 cm length. It has been manufactured in PEEK (PolyEtherEther-Ketone) and Suprasil® optical windows have been integrated for the tagging access. Image processing allows extraction of velocity profiles for a pressure driven steady flow of argon through this channel. These profiles are compared to the theoretical profiles of laminar flows and the accuracy of the method is discussed. The MTV potential for the analysis of internal gaseous flows is commented on, with a discussion on perspectives for velocity measurement in rarefied flows and direct access to slip velocity at the walls.

1. Introduction

Fluidic microsystems working with gases experienced an important development over the last years. An interesting effort of research groups concerned the development of models for the slip flow regime, frequently encountered in gaseous microflows. These models based on different boundary conditions need to be compared to experimental data in order to discuss their accuracy and limits of applicability. The experimental data on gas flows available in the literature, however, mainly concern mass flowrate measurements [1-4]. There is a crucial need for local data on pressure, velocity and concentration.

Particle Image Velocimetry (PIV) is widely used at macro-scales for flow characterization. At micro-scales (µPIV), three main aspects have to be taken into account [5]. First, as the spatial resolution is in the order of micrometers, particles of 100-300 nm are therefore needed. Second, due to their very small size, particles are submitted to the influence of the Brownian motion, which makes difficult the detection of a particle pattern by cross- or auto-correlation in an interrogation window. Third, in µPIV the whole flow volume is illuminated and a microscope lens (with high numerical aperture and narrow depth of field) is necessary to analyse a thin slice of the domain in which the particles image is sharp. This volume illumination increases the background noise due to out-off focus particles, especially if the particle density is high, which is generally the case in µPIV.
The use of Molecular Tagging Velocimetry (MTV) for local gas microflows analysis is a relatively recent step. MTV is an interesting alternative to Particle Image Velocimetry (PIV) to get local velocity information. However, MTV also presents some limitations at micro scales which will be highlighted in this paper.

MTV is a non-intrusive technique for measuring fluid velocities. It is based on the properties of gaseous molecules, such as acetone or biacetyl, used as tracers. These molecules can experience relatively long lifetime luminescence once excited by the appropriate laser wavelength. This luminescence exhibits two steps. First, fluorescence appears very rapidly after the laser excitation: its lifetime duration is in the order of a few nanoseconds. It is followed by phosphorescence, with lifetime duration of the order of hundred microseconds. A pulsed laser is used to tag these molecules (along a grid pattern or a single line in the simplest configuration) and a luminescent signal is then emitted. In the example illustrated in figure 1, the single line is detected at two successive times: a straight line is acquired a few nanoseconds after the laser beam ($t = t_0$) and a distorted one after a delay ($t = t_0 + \Delta t$). The velocity profile is determined from the Lagrangian displacement between the two images during the time $\Delta t$, assuming a flow parallel to the channel axis.

![Figure 1: MTV principle: (a) luminescent molecules have been added to the gas flow, (b) molecules are excited with the UV laser beam, (c) fluorescence or early phosphorescence signal is detected (initial tagged line), (d) phosphorescence signal is detected after a $\Delta t$ delay (distorted line).](image)

MTV does not require any seeding particle and its resolution is defined by the laser beam diameter (the lowest value cited in the literature is in the order of a few tens of µm [6]). This technique is efficient for liquid mini [7-9]- and micro-flows [10-12]. For gas flows, MTV has been successfully used at millimetric scale for external gas flow configurations [13-17]. It is considered as an excellent candidate for the analysis of rarefied gas flows in internal configurations.

MTV by direct phosphorescence is implemented in this paper. This specific MTV technique was already applied in external gas flows at millimetric scales [6]. The goal of the present work is to demonstrate the potentiality of MTV for the analysis of internal gas flows in mini-channels. It is a first step toward the visualisation of velocity profiles in rarefied flow conditions, and the direct measurement of velocity slip at the channel walls.

The experimental setup as well as the tracer properties are presented in Section 2, followed by a description of the flow cell. Then, details about data acquisition are provided. Finally, first experimental data are presented and compared to theoretical velocity profiles.

2. Experimental setup

2.1. Overall setup

A global view of the experimental setup is presented in figure 2. This setup consists of a MTV acquisition system (figure 3) and a flow/ pressure control system (figure 4).
The MTV setup includes tagging and detecting elements (figure 2), as well as a data processing unit. Tagging is made by a Quantel Twins Brilliant Nd: YAG laser (5) with a wavelength of 266 nm. The laser generates 4 ns pulses, and provides a beam, 6 mm in diameter, which is focused with an optical system in order to generate a microbeam of 35 µm diameter, which will tag acetone molecules. The available energy of the laser is reduced from 30 mJ to 1 mJ, in order to avoid damaging the channel walls due to the high energy density of the focused beam. A La Vision laser guiding arm (2) is used to safely guide the laser beam perpendicularly to the channel (3). Detection is insured by an acquisition system (figure 3) which consists in a La Vision 12-bit progressive scan camera with CCD detector (4) coupled to a 25 mm intensified relay optics -IRO- (1). The CCD has 1376 × 1040 pixels. It is operating at a frequency of 10 frames per second. The IRO is equipped with a S20 type photocathode with a P46 phosphor. An IRO controller unit (7) allows the control of the IRO timing setup, in terms of delay, gate and gain. The detected signal is sent to a computer (10) equipped with an acquisition and processing software. The whole system is externally synchronized by a programmable timing unit -PTU- (8). A protective tent is added around the channel during the acquisition to prevent possible signal perturbation by external light sources; it allows a significant improvement of the image quality by increasing the signal to noise ratio.

A gas circuit (9) supplies the channel with argon (6). It is presented in figure 4. A small intermediate reservoir is used to create a buffer volume for smoothing possible pressure fluctuations. Acetone molecules are added to the flow by bubbling argon gas through a liquid acetone bath. Argon seeded with acetone molecules enters the channel, equipped with capacitive pressure gauges (CGA.
and CGB) at its inlet and outlet. The downstream pressure can be reduced by the vacuum pump 2 connected to the channel outlet and controlled by regulating valves.

![Diagram of gas circuit and pressure control.]

**Figure 4:** Gas circuit and pressure control.

### 2.2. Acetone properties

Also called 2-propanone, acetone (CH\(_3\)-CO-CH\(_3\)), which molar weight is 58 g/mol, absorbs UV light between 200 and 320 nm (figure 5a), with an absorption peak at 275 nm [18]. With a wavelength of 266 nm, a quadrupled Nd-YAG laser is appropriate for acetone molecules excitation.

Figure 5(b) represents the normalized intensity of acetone luminescence once excited with a 266 nm wavelength. Both fluorescent and phosphorescent signals are in the blue spectrum, between 300 and 550 nm with a peak around 400 nm [18]. Phosphorescence, however, exhibits a much lower intensity; about 10 times lower than for fluorescence. As fluorescence and phosphorescence are observed in the same wavelength range, both signals can not be separated by simple spectral filtering. They appear, however, at different times after excitation. In addition, fluorescence and phosphorescence lifetime durations are typically 4 ns and 200 μs [19], respectively. These values are sensitive to pressure and temperature conditions. Additional properties of acetone can be found in [18-20].

![Acetone absorption spectrum; (b): Acetone emission spectrum: fluorescence (solid blue line) and phosphorescence (dashed red line), after [18].](a) ![Normalized photoc density](b)

**Figure 5 (a):** Acetone absorption spectrum; **(b):** Acetone emission spectrum: fluorescence (solid blue line) and phosphorescence (dashed red line), after [18].

Phosphorescence is strongly quenched by oxygen. Consequently, MTV by direct phosphorescence is hardly implementable for the analysis of gases which contain oxygen [18].
3. Flow cell
The flow cell (figure 6) has a multi-layer channel design: the main part of the device is in Polyether Ether Ketone (PEEK), mechanically machined by milling and polishing steps. The channel is 200 mm in length and has a cross section of 1 mm × 5 mm.

Two Suprasil® lenses are integrated on the PEEK block for the laser beam access. Suprasil® is a fused silica grade material transparent to UV. It has been chosen among other UV transparent materials because of its low luminescence intensity. A comparative study of this intensity (I) over time is presented in figure 7 for samples of fused quartz (TSC3®), Sapphire and fused silica (Suprasil® and Dynasil® grades). It highlights the high reemission level of TSC3® and Sapphire compared to fused silica. This strong luminescence of the walls was able to mask the phosphorescence of the acetone molecules.

![Figure 6: Channel flow.](image)

![Figure 7: Reemission decay curves of different materials after excitation by 266 nm wavelength light. (a): TSC3® (blue diamond) and Sapphire (purple square); (b): Suprasil® (blue diamond) and Dynasil® (purple square).](image)
Suprasil® lenses (A) are located at the inlet and at mid-length of the channel (figure 8). Two supplementary lenses (B) are also integrated in front of the first ones to allow transmission of the laser beam, and limit absorption and reflection at the wall. Two Borofloat® plates permit visualisation perpendicularly to the laser beam. Borofloat® is a grade of glass provided by Zell Quartz Glass industry. These plates are sealed with acetone resistant o-rings. Type K thermocouples are added along the channel wall to provide thermal information. Suprasil® lenses and thermocouples are flush mounted in order not to disturb the flow (figure 8).

![Figure 8: Thermocouples and Suprasil® lenses alignment.](image)

The set of the flow cell layers are clamped between two PEEK sheets (figure 6). Swagelok® connectors are used for the flow inlet and outlet accesses. They are parallel to the streamwise direction and connected to the whole setup via two small chambers equipped with pressure capacitive gauges (figure 9).

![Figure 9: Flow cell.](image)

4. Data acquisition and analysis

4.1. Image acquisition and processing

Acquisition parameters (IRO delay, gain, gate, camera exposure time...) are controlled with Davis® acquisition software and checked with a 2-channel oscilloscope. Two acquisition methods, the “standard averaging” and “on chip integration” techniques, are investigated herein.
4.1.1. Standard averaging technique: In this technique, a set of instantaneous images - one image per camera exposure time and per laser pulse - is captured and averaged. A simplified timing scheme is presented in figure 10 to illustrate this technique.

\[ t_0 \] and \[ t_1 \] represent the time of the initial and deformed tagged lines acquisitions, respectively. Images at \( t_0 \) and \( t_1 \) are taken separately, i.e. in two separate camera exposure times. For the images presented in figure 11, the camera exposure time is set to 2000 µs. The signal acquisition duration on the CCD camera is controlled by the intensifier activation time (IRO gate) \( G \) set to 1 µs and \( t_1 = 100 \mu s \). In order to obtain the image in figure 11 (b), a set of 10000 instantaneous images is captured. These images are then post-processed with a statistical average followed by a Gaussian smoothing operation. Post processing increases the signal to noise ratio which is very poor for a single image (figure 11-a). The average of a set of images (figure 11-b) exhibits a high signal/noise ratio and therefore allows extracting velocity profiles from the deformed line acquisition. The acquisition of such a high number of images requires, however, significant time duration (about 40 min for 10000 images), during which a precise control of pressure conditions is required.

![Figure 10: Timing scheme of a single image acquisition for the standard averaging technique.](image)

4.1.2. On chip integration technique: In this method, the CCD exposure time is long enough to capture the emission of more than one laser pulse. This helps to increase the signal to noise ratio, as several single luminescent signals are cumulated in one CCD exposure, while being affected by readout noise only once. The corresponding timing diagram is presented in figure 12.

![Figure 11 (a): One instantaneous image; (b): Averaged image from a set of 10000 images.](image)
The signals obtained with both techniques are compared in figure 13.

![Figure 12: On chip integration timing diagram.](image)

**Figure 13 (a):** On chip integration signal, averaging of 1000 images with 50 pulses integration per image; **(b):** Standard averaging technique, averaging of 10000 images with 1 laser pulse per image.

Figures 13-a and 13-b are not taken with exactly the same flow conditions neither at the same time delay, which explains the small deviations in the deformed profile. It can be observed that the on chip integration technique gives a better signal quality. In addition, the acquisition time is reduced with this technique and a constant operating pressure is then more easily controlled.

4.2. Analytical solution

First experimental velocity profiles obtained with the MTV technique are compared with the analytical solution of a laminar fully developed gas flow through rectangular channel, which can be written in a dimensionless form [21]:

\[
V(x, y) = \sum_{n=1,\text{odd}}^{\infty} \sum_{m=1,\text{odd}}^{\infty} V_{n,m} \sin(n\pi x) \sin\left(\frac{m\pi y}{\beta}\right)
\]

(1)

with

\[
V_{n,m} = \frac{\pi^2}{4mn(\beta^2n^2 + m^2)} \sum_{i=1,\text{odd}}^{\infty} \sum_{j=1,\text{odd}}^{\infty} \frac{1}{i^2 j^2 (\beta^2i^2 + j^2)} = \frac{1}{A(\beta)mn(\beta^2n^2 + m^2)}
\]

(2)
In Eq. (2), \( A(\beta) \) is a function of the aspect ratio \( \beta \) of the cross-section and can be accurately approximated by the polynomial \( 0.5059 - 0.3022 \beta - 0.0642 \beta^2 + 0.0747 \beta^3 \) \([21]\); \( i, j, n \) and \( m \) are summation integer indices; \( x \) and \( y \) are dimensionless rectangular Cartesian coordinates; \( \nu_{i,m} \) is defined by Eq. (2).

5. First results and discussion

A typical MTV result is presented in figure 14 with a superposition of initial and deformed tagged lines. The deformed tagged purple line is taken with a time delay \( \Delta t = 99.49 \, \mu s \) after the initial green line. The spot observed at the bottom of the green line is due to a high energy level and a reflexion of the luminescence at the wall.

The decrease in phosphorescence intensity level with time was expected, as well as the phosphorescence lifetime duration (about 150 \( \mu s \)) which is comparable with data from the literature \([18]\). A local temperature increase can result in a phosphorescence time decrease, but this possible local effect is at the moment difficult to quantify. The temperature, which is measured at 3 different

![Figure 14: Superposition of initial and deformed MTV images in the median plane of the channel at equal distance from lateral walls.](image)

![Figure 15: Intensity profiles of the signal at the channel centreline: a) initial tagged line at \( t_0 = 0.51 \, \mu s \); b) deformed tagged line at \( t_1 = 100 \, \mu s \).](image)
locations along the channel, is constant during the recording time (figure 16), but the presence of a hot spot is possible in the tagged region and could influence the duration of phosphorescence.

Figure 16: Temperature evolution at the channel wall.

The obtained deformed tagged line is presented in figure 17. It is successfully superposed to the theoretical displacement profile calculated from the velocity given by Eq (1) for $\Delta P = 190$ Pa and $\Delta t = 99.49$ µs.

Figure 17: Superposition of deformed tagged line and theoretical displacement profile for $\Delta P = 190$ Pa and $\Delta t = 99.49$ µs. Red edges correspond to channel walls location.

Velocity profile in the median channel plan (figure 18) is extracted from the experimental data presented in figure 17 and compared to the theoretical Poiseuille flow profile. The uncertainties on some parameters are taken into account. For the channel height, the uncertainty $\Delta h/h = \pm 3\%$. For the measured pressure, the uncertainty $\Delta P/P = \pm 0.4 \%$. For the viscosity, the uncertainty $\Delta \mu/\mu = \pm 0.13 \%$. Considering a plane flow between two parallel plates (in the middle of the channel, far from the walls), the total uncertainty is written as $\frac{\Delta U}{U} = 2\frac{\Delta h}{h} + \frac{\Delta P}{P} + \frac{\Delta \mu}{\mu} = \pm 6.53 \%$. 
Figure 18: Velocity profile in a cross-section: experimental data (blue) and velocity profile in rectangular duct (pink, red and green) obtained for $\Delta P = 190$ Pa.

In figure 18, the theoretical velocity profiles in a rectangular channel are plotted taking into account this total uncertainty: respectively $+6.53\%$ for the red and $-6.53\%$ for the green dashed lines. Small deviations are observed between experimental and theoretical profiles. The dispersion observed on the experimental data is mainly due to the technique used to extract these data. The displacement of the tagged molecules has been deduced from the position of the peak on a series of intensity curves like the one shown in figure 15-a, corresponding to a scan of the image along the $x$-direction at different $y$-positions, i.e. at different depths in the channel. Work is currently in progress to improve this data extraction, which maybe requires an initial smoothing of the intensity curves.

6. Conclusion and perspectives
A Molecular Tagging Velocimetry setup for gas flow analysis has been developed. An original flow cell of $1\, \text{mm} \times 5\, \text{mm}$ cross-section with integrated temperature sensors has been manufactured. First promising experimental results have been obtained. The extracted velocity profiles are successfully compared to velocity profiles of a compressible Poiseuille flow in a rectangular duct. The potential of the MTV technique for the analysis of internal gas flows is therefore demonstrated. As the final goal of the work is to access velocity profiles in the slip flow regime, next tests will concern flows at lower pressures. In addition, the study of the temperature phosphorescence dependence is foreseen. This could be a first step toward Molecular Tagging Thermometry (MTT) which would be an interesting tool for the analysis of heat transfer in the slip flow regime, for which there is a crucial need in the literature [22].

Acknowledgment
This research obtained financial support from the European Community’s Seventh Framework Program (FP7/2007-2013) under grant agreement no 215504 and from the ‘Fédération de Recherche Fermat’, FR 3089, Toulouse, France. Dr. Sébastien Cazin and Dr. Emmanuel Cid are acknowledged for valuable discussion.

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