Molecular tagging velocimetry for confined rarefied gas flows: Phosphorescence emission measurements at low pressure

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ABSTRACT

Rarefied gas flows have a central role in microfluidic devices for many applications in various scientific fields. Local thermodynamic non-equilibrium at the wall-gas interface produces macroscopic effects, one of which is a velocity slip between the gas flow and the solid surface. Local experimental data able to shed light on this physical phenomenon are very limited in the literature. The molecular tagging velocimetry (MTV) could be a suitable technique for measuring velocity fields in gas micro flows. However, the implementation of this technique in the case of confined and rarefied gas flows is a difficult task: the reduced number of molecules in the system, which induces high diffusion, and the low concentration of the molecular tracer both drastically reduce the intensity and the duration of the exploitable signal for carrying out the velocity measurements. This work demonstrates that the application of the 1D-MTV by direct phosphorescence to gas flows in the slip flow regime and in a rectangular long channel is, actually, possible. New experimental data on phosphorescence emission of acetone and diacetyl vapors at low pressures are presented. An analysis of the optimal excitation wavelength is carried out to maximize the intensity and the lifetime of the tracer emission. The experimental results demonstrate that a low concentration of about 5–10% of acetone vapor excited at 310 nm or of diacetyl vapor excited at 410 nm in a helium mixture at pressures on the order of 1 kPa provides an intense and durable luminescent signal. In a 1-mm deep channel, a gas flow characterized by these thermodynamic conditions is in the slip flow regime. Moreover, numerical experiments based on DSMC simulations are carried out to demonstrate that an accurate measurement of the velocity profile in a laminar pressure-driven flow is possible for the rarefied conditions of interest.

1. Introduction

In the last two decades, micro-electro-mechanical systems (MEMS) have received a lot of attention due to their appealing properties of low volume and weight, and for their possible applications in a very wide variety of scientific fields. Specifically in the gas microfluidic sector, several interesting applications have recently been developed. It is the case of micronozzles for space applications [1], micro-actuators for aeronautical applications [2], micro heat exchangers [3], and Knudsen pumps [4], to name just a few. The growing interest towards these micro-devices has brought the scientific community to dedicate great research effort towards heat and mass transfer analysis in gas flows at microscale.

The size reduction in micro gas systems amplifies the thermodynamic disequilibrium of the flow as a consequence of the increased gas rarefaction. This phenomenon results from a decrease in the number of intermolecular collisions inside the control volume. The Knudsen number, $Kn = \lambda/L_c$, where λ is the mean free path and L_c is the characteristic size of the system, identifies the degree of gas rarefaction. Most of the microfluidic devices works in the slip flow regime, with Kn in the range $[10^{-3}; 10^{-1}]$, which is to be considered as a slightly rarefied gas flow regime. This regime can be reached either in a microfluidic device, where L_c is low, or in a bigger system at low pressure, which results in a high mean free path λ. In this rarefaction regime, the continuum representation of the flow is still acceptable for fluid particles that are far enough from the system's solid boundaries, i.e. at a distance larger than the well-known Knudsen layer thickness which is considered to be of the order of λ. However, for those systems that involve wall-gas interactions, e.g., in micro channels or in channels at low average pressure, the collisions between the wall surfaces and the

gas molecules gain importance with respect to intermolecular collisions, thus producing a thermodynamic non-equilibrium state of the gas in the vicinity of the wall. At a macroscopic level, this produces a velocity slip and a temperature jump at the wall [5], i.e., discontinuities of kinematic and thermodynamic parameters between fluid and surface. Therefore, even if the Navier-Stokes-Fourier equations still hold, specific boundary conditions must be introduced to take into account these discontinuities at the wall. Various forms of boundary conditions for both continuum medium and kinetic theory [6] derived models have been proposed in the literature. All of them depend on accommodation coefficients that vary as a function of the wall properties and gas species, and which determination is still a difficult task. Experimental data are required to evaluate these coefficients and to validate the proposed models.

Most of the experiments performed on rarefied gas flows in channels indirectly analyze the effects given by the velocity slip and temperature jump at the wall by measuring global quantities, such as mass flow rate, inlet and outlet pressures and temperatures [7–14]. To the best of our knowledge, there are no experimental data in the literature that provide local analysis of the rarefaction effect at the wall.

One of the most widely used techniques for local investigation of the velocity field in both liquid and gas flows is the particle image velocimetry (PIV). Its application to microfluidic systems is known as μPIV. Although this technique can provide accurate data related to liquid flows in micro-channels [15–17], the application of μPIV has been very limited up to now in gas micro flows [18,19]. The main problem is related to the tracer particle size. Beside the technological difficulties in generating particles with a diameter less than 1 μm, the particle size should be small enough to guarantee that the particles faithfully follow the fluid flow and big enough to reduce Brownian motion noise [20] that makes the cross-correlation operation inaccurate. For these reasons, μPIV has never been applied for velocity measurements in rarefied gas flows.

Our team has developed a molecular tagging velocimetry technique with the purpose of directly quantifying the slip velocity at the wall and measuring the overall velocity profile for a pressure-driven rarefied gas flow in a channel of rectangular section.

The molecular tagging velocimetry (MTV) is an optical technique with low intrusiveness. Differently from PIV where the tracer is made of particles, this alternative velocimetry technique is based on the exploitation of a molecular tracer able to emit light for a certain duration after having been excited by a UV-light source. Among all the possible versions of this technique, 1D-MTV by direct phosphorescence of acetone (CH_3COCH_3) or diacetyl ($CH_3(CO)_2CH_3$) vapors has been chosen for the present work. The basic principle of this technique is depicted in Fig. 1. The velocity profile is deduced from the streamwise displacement of the tracer molecules initially tagged by the laser along a line perpendicular to the gas flow direction.

1D-MTV by direct phosphorescence is the simplest and the most straightforward version of MTV, since it requires only one laser system, in contrast to other implementations, such as RELIEF [21] or PHANTOMM [22] techniques, which require more than one photon source or cannot be applied to gas flows. Laser sources with high repetition rates can be useful for velocimetry in unsteady flows. For application to

steady gas flows, a frequency of the order of 10 Hz provided by common flashlamp-pumped lasers is high enough to accurately measure velocity fields.

1D-MTV by direct phosphorescence has already been applied to non-rarefied external supersonic or hypersonic turbulent gas flows [23,24], to rarefied supersonic jets [25] and to non-rarefied gas flows in millimetric channels [26,27]. However, some difficulties have prevented until now the successful application of this technique to confined rarefied gas flows. Since the laser beam can hardly be smaller than about 30 μm for technological reasons, the height of the channel is constrained to be not smaller than about 1 mm, in order to keep a reasonable spatial resolution. Consequently, Knudsen numbers corresponding to the targeted slip flow regime can only be reached by decreasing the average pressure of the gas-tracer mixture.

Our research team has already made progress in the direction of applying MTV in rarefied conditions. Samouda et al. [26] have demonstrated that the technique can provide good results in a millimetric rectangular channel for a non-rarefied gas flow at atmospheric pressure and ambient temperature. However, they noticed that a deduction of the velocity profile by assuming it was simply homothetic of the displacement profile resulted in an artificial velocity slip at the wall, which was totally unexpected for the considered Knudsen numbers. Subsequently, Frezzotti et al. [28] explained this unexpected phenomenon at the wall as a consequence of a combined effect of advection and molecular diffusion of the tracer in the background gas flow. Moreover, the same authors proposed a numerical method based on a simple advection-diffusion equation that was able to correctly reconstruct the velocity profile from the displacement profile. By means of the Direct Simulation Monte Carlo (DSMC) method, it was possible to numerically verify the existence of a displacement slip at the wall caused by the advection-diffusion mechanism and not linked to a velocity slip at the wall. A reconstruction method of the velocity profile from the displacement profile was developed and validated with numerical experiments. Si Hadj Mohand et al. [27] successfully applied this reconstruction method on MTV data in a millimetric channel and correctly extracted the velocity profile at atmospheric and sub-atmospheric pressures down to a minimum average pressure of 50 kPa. At this pressure level in a 1-mm deep channel, the flow is still in a non-rarefied regime.

Lower pressures reduce, however, the tracer molecule concentration and increase the molecular diffusion. The combination of these two effects drastically decreases the phosphorescence signal intensity as well as its lifetime. Therefore, it is necessary to experimentally explore the phosphorescence emission of different molecular tracers and look for a physical condition that makes the application of the 1D-MTV to the rarefied case feasible.

For that purpose, the present work is intended to show new results in terms of acetone and diacetyl phosphorescence as a function of total pressure, molecular tracer concentration, excitation wavelength, and gas species, in low pressure conditions. These experimental data reveal which Knudsen numbers can be achieved in a pressure-driven flow while still having a durable and visible phosphorescence signal from the tracer. In this work, the Knudsen number,
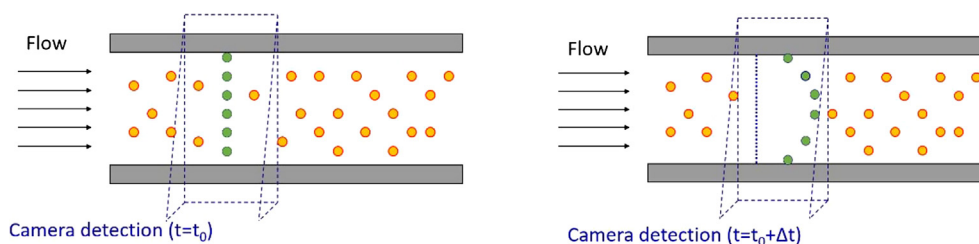
$$Kn = \lambda/L_y, \tag{1}$$

Fig. 1. Basic principle of 1D-MTV by direct phosphorescence, for a gas flowing in a plane channel from left to right.