MIGRATE-154814

ACETONE AND DIACETYL PHOSPHORESCENCE AT LOW PRESSURES FOR MOLECULAR TAGGING VELOCIMETRY IN CONFINED RAREFIED GAS FLOWS

Dominique Fratantonio, Varun Yeachana, Marcos Rojas-Cardenas, Christine Barrot, Stéphane Colin*
Institut Clément Ader (ICA), Université de Toulouse, CNRS, INSA, ISAE-SUPAERO, Mines-Albi, UPS, Toulouse France
stephane.colin@insa-toulouse.fr

KEY WORDS

Acetone phosphorescence, diacetyl phosphorescence, molecular tagging velocimetry, rarefied gas.

ABSTRACT

In the last twenty years, the development of miniaturized electromechanical systems (MEMS) has exponentially increased, as a result of the big technological progress on the micro-fabrication methods. The scientific research followed this trend by spending a lot of resources on the theoretical, numerical, and experimental analysis of fluidic MEMS for understanding and controlling the physics governing them. Among the different possible types of fluidic MEMS, there is a group of micro-devices which functioning requires the control of gas micro-flows, such as micro-actuators for aeronautical applications and Knudsen pumps [1,2].

Even if a lot of research effort has been already dedicated to the understanding of rarefied gas flows, until now most of the experimental studies available in the literature provide analyses based on the measurement of global quantities, typically the mass flow rate, and the pressure or temperature differences [3,4]. This approach allows to indirectly analyze the microscopic effects that the velocity and temperature discontinuities at the wall generate [5]. Our interest in the application of the molecular tagging velocimetry (MTV) to the case of rarefied and confined gas flows is then motivated by the lack of experimental local information. To the best of our knowledge, there is no evidence in the literature of experimental data that provide a direct measurement of the slip velocity at the wall characterizing pressure or thermally-driven rarefied gas flows. For these reasons, this work aims to the application of the 1D-MTV technique, the simplest version of this technique, to gas flows in the slip regime in a channel of rectangular section, by exploiting either acetone or diacetyl vapor as molecular tracer.

The Poiseuille gas flow is considered as a benchmark case. In order to experimentally obtain the typical parabolic velocity profile, the channel width must be large enough with respect to the channel height. The degree of gas rarefaction is determined by comparing the mean free path with the smallest dimension of the system. Therefore, for attaining a Knudsen number in the range [0.1 – 0.01], namely in the slip flow regime, either the height of the channel or the pressure must be reduced. The reduction of the channel height, however, is limited by the fact that the tagged line size is lower bounded. Actually, the laser beam diameter provided by most of the laser systems cannot be reduced to less than

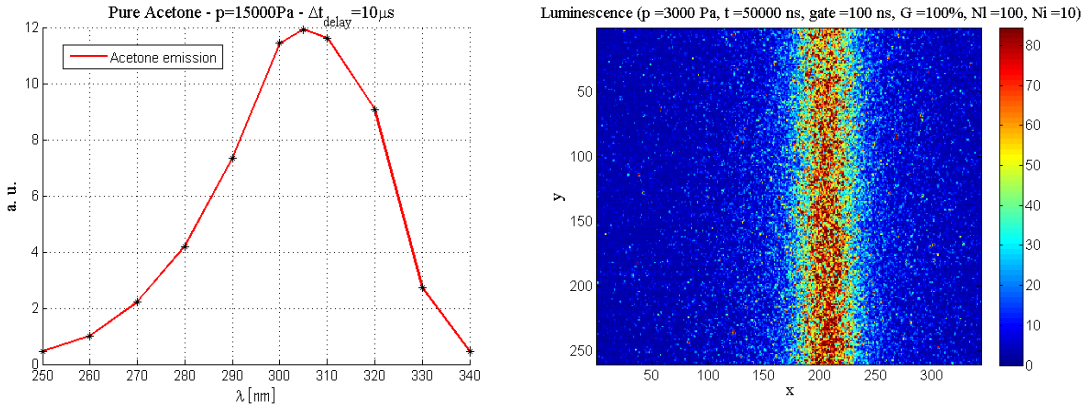
about 30 μm. Therefore, in order to achieve the slip flow regime, it is necessary to reduce the gas average pressure.

In our past work, it was showed that the acetone (CH_3COCH_3) vapor excited with a laser beam at a wavelength of 266 nm does not emit enough light for its use as a molecular tracer for the application of the MTV, at least with the current intensifier and camera available on the market. Even if the excitation is made at a wavelength very close to the pic of acetone absorption, which is 271 nm [6], the phosphorescence emission lasts only for some microseconds when the acetone partial pressure is lower than 5 kPa. As a consequence of low acetone molecule concentration, molecular diffusion of the tagged molecules and limitations on the excitation energy, the resulting phosphorescence signal is too weak. By considering a channel of 1 mm in height and a helium-acetone flow with 5% of acetone vapor, the average pressure should be at least 1600 Pa for obtaining a Knudsen number of 0.01, i.e. for having a gas flow in the slip regime. This thermodynamic condition requires an acetone partial pressure of only 80 Pa. Therefore, it is evident that acetone vapor excited at 266 nm cannot be employed as a molecular tracer for direct measurement of slip velocity at the wall.

However, we discovered that changing the excitation wavelength from 266 nm up to 310 nm can drastically increase the duration and the intensity of the phosphorescent signal. Even if the photon absorption of the acetone is lower at 310 nm than at 266 nm, the increase of the phosphorescence quantum yield, i.e. the percentage of excited molecules that finally phosphoresce, is higher than the decrease of the absorbing molecules number. According to some works belonging to chemistry's literature, the intramolecular transitions that occur for this phenomenon to happen are actually complex. The increase in light emission could indeed be due to the presence of diacetyl, which forms following molecular dissociation of the acetone caused by the 310 nm excitation [7, 8].

Furthermore, the possibility to use diacetyl ($CH_3(CO)_2CH_3$) vapor as a possible molecular tracer by analyzing the intensity and the duration of its phosphorescence has been investigated. The direct excitation of diacetyl molecules produces even more intense and more durable phosphorescence than the acetone excited at 310 nm. As a matter of fact, the phosphorescence quantum yield of acetone and diacetyl are estimated to be 1.8 % and 15 %, respectively [6]. Although diacetyl provides higher signal intensity, the interest in exploring the possibility to use acetone vapor is motivated by the fact that diacetyl is more toxic. Moreover, it has been experimentally observed that diacetyl presents more wall adsorption problems than acetone, which introduce further difficulties in controlling the experimental conditions.

In this work, experimental data on phosphorescent emission of acetone and diacetyl vapor as a function of the excitation wavelength are presented (Figure 1a). Once the optimal excitation wavelength is found, the phosphorescence lifetime of both molecular tracers is investigated at low pressures and in helium mixtures. For example, Figure 1b shows the phosphorescent emission of diacetyl in helium at 3 kPa. In the light of the presented experimental data, the possible use of acetone and diacetyl vapor for a successful application of the MTV to rarefied gas flows in a rectangular channel is discussed.



(a) (b)

Figure 1: (a) phosphorescent emission of acetone vapor at 15 kPa as a function of excitation wavelength. The signal intensity results from the collection of photons emitted during 100 ns, and 10 μs after the laser excitation. The data are normalized with respect to the intensity measured at an excitation wavelength of 260 nm; (b) phosphorescent emission of diacetyl in helium at p = 3 kPa with a diacetyl molar fraction $\chi \cong 10$ %. The image represents the light emission collected during 100 ns, and 50 μs after the laser excitation.

Acknowledgements

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.

References

[1] An, S., Gupta, N. K., and Gianchandani, Y. B. (2014). A Si-micromachined 162-stage two-part Knudsen pump for on-chip vacuum. *Journal of Microelectromechanical Systems* **23**: 406-416.

[2] Cattafesta, L. N., and Sheplak, M. (2011). "Actuators for active flow control". *Annual Review of Fluid Mechanics* **43**: 247-272.

[3] Pitakarnnop, J., Varoutis, S., Valougeorgis, D., Geoffroy, S., Baldas, L., & Colin, S. (2010). A novel experimental setup for gas microflows. *Microfluidics and Nanofluidics* **8**(1): 57-72.

[4] Silva, E., Rojas-Cardenas, M., and Deschamps, C. J. (2016). Experimental analysis of velocity slip at the wall for gas flows of nitrogen, R134a, and R600a through a metallic microtube. *International Journal of Refrigeration* **66**: 121-132.

[5] Colin, S. (2012). Gas microflows in the slip flow regime: a critical review on convective heat transfer. *Journal of Heat Transfer* **134**(2): 020908.

[6] Lozano, A., Yip, B., & Hanson, R. K. (1992). Acetone : a tracer for concentration measurements in gaseous flows by planar laser-induced fluorescence. *Experiments in Fluids* **13**: 369-376.

[7] Almy, G. M., and Anderson, S. (1940). Lifetime of fluorescence in diacetyl and acetone. *The Journal of Chemical Physics* **8**: 805-814.

[8] Heicklen, J., and Noyes Jr, W. A. (1959). The photolysis and fluorescence of acetone and acetone-biacetyl mixture. *Journal of the American Chemical Society* **81**: 3858-3863.