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## MEASUREMENTS OF DIFFUSION COEFFICIENT AND KINETIC DIAMETER OF DIACETYL AND ACETONE VAPOR VIA MOLECULAR TAGGING

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### KEY WORDS

Diffusion, Kinetic diameter, Molecular tagging, Photoluminescence, Acetone, Diacetyl.

### ABSTRACT

#### Introduction

In recent years, gas microflows have gained increasing interest due to their specific microscale behavior and for their appealing microfluidic applications. Microdevices involving gas microflows, such as micro heat exchangers [1], Knudsen pumps [2], artificial lungs [3], and pressure sensors [4], cover a large spectrum of industrial and scientific domains (transport, health, energy, environment, aerospace, etc....).

When the dimensions of fluidic devices are reduced, the continuum medium assumption can become invalid for gases, as rarefaction effects become significant and thus the classical no-slip boundary conditions are no longer valid [5]. For a rarefied gas, the mean free path  $\lambda$  of the molecules is comparable or even larger than the characteristic length  $L_c$  of the microdevice. For slightly rarefied gas flows, typically for a Knudsen number  $Kn = \frac{\lambda}{L_c}$  between  $10^{-3}$  and  $10^{-1}$  corresponding to the so-called slip-flow regime, a local thermodynamic disequilibrium in the Knudsen layer at the wall leads to a velocity slip and a temperature jump that impact the macroscopic velocity and temperature distributions within the flow. In addition, as the number of gas intermolecular collisions is reduced, molecular diffusion has a more significant effect on heat and mass transfer.

A better understanding of the transport phenomena at the microscale is necessary for a precise design and optimization of fluidic microsystems involving gases. Direct measurements of the slip

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velocity and temperature jump at the wall are necessary for the validation of the numerical models, and more specifically in respect to the values of the associated accommodation coefficients [6-8]. However, most of the experimental studies published in the literature focus on measurements of global quantities, such as the mass flow rate, or pressure and temperature differences between the inlet and outlet of microchannels. Given the reduced dimensions of the investigated domain, it is very challenging to obtain accurate measurements of velocity and temperature distributions in a gas microflow, especially close to the wall and in the Knudsen layer.

### Molecular tagging

This scientific objective pushed our group to develop a molecular tagging (MT) technique for local velocimetry and thermometry application [9; 10]. Molecular tagging is an optical, and low-intrusive measurement technique, which exploits the laser-induced photoluminescence of specific tracer molecules seeding the gas flow under investigation [11-15]. Acetone and diacetyl vapors are two of the most widely used tracers for single-laser MT in gaseous environment.

The molecular tagging velocimetry (MTV) technique consists in recording the displacement of the luminescent tracer molecules during a short time interval, in the micro-to-millisecond range. A good spatial resolution requires operating experiments in devices with millimetric characteristic lengths. In order to reach the slip flow regime of interest at this scale, the gas pressure must be low enough, typically of the order of 0.1-1 kPa. The MTV technique has been successfully implemented to measure velocity profiles in a rectangular mini-channel [11; 13; 14], and to observe a slip velocity at the wall in a helium flow seeded with acetone vapor, at a pressure of the order of 1 kPa and a Knudsen number of the order of  $10^{-2}$  [11; 13; 14].

MTV velocity measurements are carried out by tracking the displacement of a tagged line of molecules between a time interval. However, the combined effect of molecular diffusion and velocity gradients prevents extracting the local flow velocity by simply dividing the local molecular displacement by the time of flight. In the case of microchannel flows, this advection-diffusion effect is known as Taylor dispersion and produces a molecular displacement at the wall even for gas flows in hydrodynamic regime, for which the macroscopic velocity at the wall is expected to be zero. As a result, the displacement profile in a 2-D Poiseuille flow exhibits a non-parabolic shape [14]. That is why a specific reconstruction algorithm [12] that takes into account the combined effect of advection and diffusion is needed, in order to obtain a correct extraction of the velocity profile from the measured displacement profile [10, 11, 12].

A reconstruction method was proposed by Frezzotti et al. and successfully applied in the continuum regime by Si Hadj Mohand et al. and by Fratantonio et al. [12] to acetone-seeded helium flows in a rectangular microchannel at relatively high pressures. Fratantonio et al. further applied this algorithm to a slightly rarefied helium gas flow, with a pressure as low as 1 kPa. At low pressures, the reconstruction algorithm failed to accurately obtain the velocity profile and the value of the slip velocity at the wall, showing that the proposed reconstruction algorithm is strongly sensitive to the diffusion coefficient  $D$  of the tracer through the background gas, and that this parameter needs to be well-characterized in order to apply the methodology. Nevertheless, the diffusion coefficient in confined flows is of difficult estimation and new experimental methods are needed in order to measure this parameter optically.

### Diffusion measurement

A successful measurement of the local flow velocity requires a correct estimation of the molecular diffusion coefficient for the employed tracer vapor. The binary diffusion coefficient calculation is based on a correct knowledge of the kinetic diameter  $d$  of acetone and diacetyl vapor, some values of which can be found in the literature (see Tab.1). The large variation in the kinetic diameter of acetone and diacetyl reported in the literature results in a large uncertainty in the molecular diffusion coefficient  $D$  when calculated by the Chapman-Enskog theory [16]:

$$D = \frac{3\sqrt{\pi}}{8} \sqrt{\frac{k_B T}{m}} \frac{1}{\sqrt{2\pi d^2 n}} \quad (1)$$

where  $k_B$  is the Boltzmann constant,  $d$  is the kinetic molecular diameter,  $n$  is the number density,  $T$  is the temperature and  $m$  is the molecular mass.

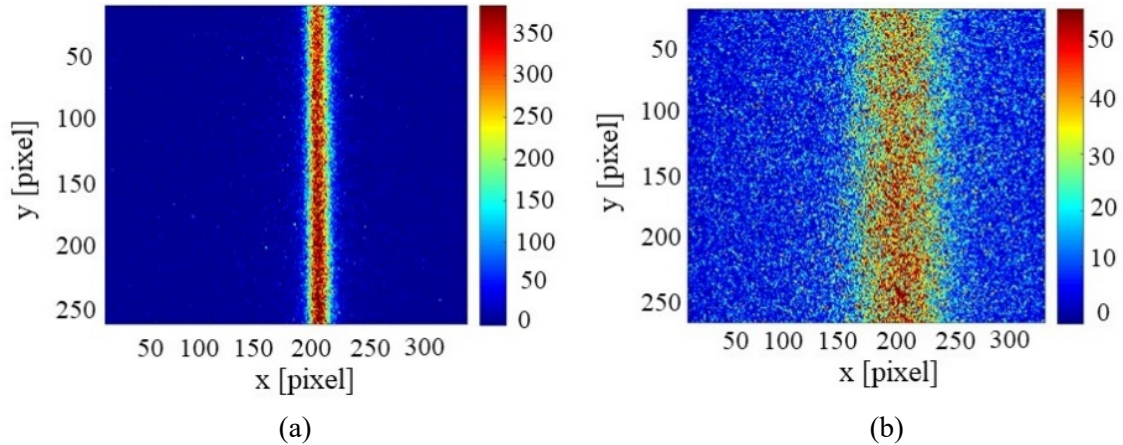
References	Kinetic diameter of acetone (pm)	Kinetic diameter of diacetyl (pm)
Hu et al. [17]	460	-
Frezzotti et al. [11]	730	-
Nadykto & Yu [18]	616	-
Bowen et al. [19]	416	-
Almy & Anderson [20]	590	590
Mirzaei et al. [21]	-	1180

**Table 1:** Kinetic diameter of acetone and diacetyl molecules published in the literature.

In this context, this work aims to provide measurements of the molecular diffusion coefficient and of the molecular kinetic diameter of gaseous acetone and diacetyl from optically acquiring the temporal evolution of the luminescence signal distribution produced by molecular tagging. The measurements were obtained for different pressures of the tracer, values ranging from close to saturation vapor pressure at room temperature to 100 Pa. As it can be observed from Fig. 1, after laser excitation, the emitted light distribution changes over time due to diffusion. It has been demonstrated in a previous work that the light distribution can be considered as a Gaussian function [15], thus the variance  $\sigma_x^2$  of the Gaussian distribution represents the diffusion effects in space. This variance should evolve linearly with time, with a slope proportional to the diffusion coefficient, as described by the Einstein's formula

$$\sigma_x^2 = 2Dt + \sigma_{x,0}^2 \quad (2)$$

where  $D$  is the diffusion coefficient,  $t$  is the time after the laser excitation,  $\sigma_{x,0}^2$  is the variance of the initial light distribution.

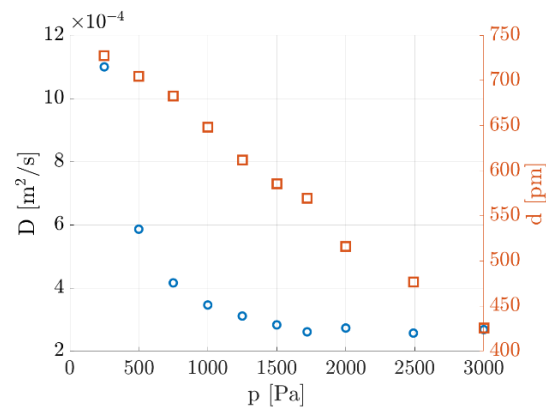


**Figure 1:** Diacetyl vapor phosphorescent signal at  $p = 1750$  Pa and  $T = 20$  °C. The scale of luminescent intensity on the right side of the figures is in arbitrary unit. Time after laser excitation: a)  $t = 0.5$   $\mu$ s; b)  $t = 300$   $\mu$ s. Figures taken from [22].

Since the Einstein relationship does not consider the totality of the intramolecular process after laser excitation, a complex analysis on the phosphorescent signal and its variation with time was realized, for the diffusion coefficient extraction from these results [20]. An optimization algorithm was developed to extract the molecular diffusion coefficient from the time-series of recorded light emissions. This algorithm is based on a numerical model of the spatial and temporal evolution of the excited tracer molecules and considers the spontaneous photon emission, the non-radiative de-excitation, and the molecular diffusion after laser excitation.

Fig.2 reports the measured diffusion coefficient and the related kinetic diameter as a function of pressure for the case of diacetyl vapor. The measured kinetic diameter decreases linearly with pressure. This result is unexpected and additional experiments are currently in progress. The mean kinetic

diameter value of diacetyl is, however, close to the value published by Almy and Anderson [18] (Tab. 1). The ongoing work will complete these data concerning larger datasets on diacetyl and acetone vapors.



**Figure 2:** Diffusion coefficient (○) of diacetyl extracted from recorded images at different pressures. Kinetic diameter  $d$  (□) calculated from the measured diffusion coefficient. Figure taken from [23].

## References and Citations

- [1] Rehman, D., Joseph, J., Morini, G. L., Delanaye, M., & Brandner, J. (2020). A Hybrid Numerical Methodology Based on CFD and Porous Medium for Thermal Performance Evaluation of Gas to Gas Micro Heat Exchanger. *Micromachines*, 11(2), 218.
- [2] Qu, C., Alphenaar, B., McNamara, S., & Walsh, K. (2020). Fabrication of Nanoporous Membranes for Knudsen Pump Using Glancing Angle Deposition. Paper presented at the 2020 IEEE 33rd International Conference on Micro Electro Mechanical Systems (MEMS).
- [3] Potkay, J. A. (2014). The promise of microfluidic artificial lungs. *Lab on a Chip*, 14(21), 4122-4138.
- [4] Javed, Y., Mansoor, M., & Shah, I. A. (2019). A review of principles of MEMS pressure sensing with its aerospace applications. *Sensor Review*, 39(5), 652-664. doi:10.1108/SR-06-2018-0135
- [5] Colin, S. (2011). Gas Microflows in the Slip Flow Regime: A Critical Review on Convective Heat Transfer. *Journal of Heat Transfer*, 134(2). doi:10.1115/1.4005063
- [6] Maxwell, J. C. (1890). *The Scientific Papers of James Clerk Maxwell (Vol. 2)*: University Press.
- [7] Ebert, W., & Sparrow, E. M. (1965). Slip flow in rectangular and annular ducts.
- [8] Arkilic, E. B., Schmidt, M. A., & Breuer, K. S. (1997). Gaseous slip flow in long microchannels. *Journal of Microelectromechanical systems*, 6(2), 167-178.
- [9] Koochesfahani, M., & Nocera, N. (2007). Molecular tagging velocimetry. *Handbook of experimental fluid dynamics*, 362-382.
- [10] Hu, H., & Koochesfahani, M. (2006). Molecular tagging velocimetry and thermometry and its application to the wake of a heated circular cylinder. *Measurement Science and Technology*, 17(6), 1269.
- [11] Fratantonio, D., Rojas-Cárdenas, M., Barrot, C., Baldas, L., & Colin, S. (2020). Velocity measurements in channel gas flows in the slip regime by means of molecular tagging velocimetry. *Micromachines*, 11(4), 374.
- [12] Frezzotti, Si Hadj Mohand, H., Barrot, C., & Colin, S. (2015). Role of diffusion on molecular tagging velocimetry technique for rarefied gas flow analysis. *Microfluidics and Nanofluidics*, 19(6), 1335-1348.
- [13] Si Hadj Mohand, H., Frezzotti, A., Brandner, J. J., Barrot, C., & Colin, S. (2017). Molecular tagging velocimetry by direct phosphorescence in gas microflows: correction of Taylor dispersion. *Experimental Thermal and Fluid Science*, 83, 177-190.
- [14] Samouda, F., Colin, S., Barrot, C., Baldas, L., & Brandner, J. J. (2015). Micro molecular tagging velocimetry for analysis of gas flows in mini and micro systems. *Microsystem Technologies*, 21(3), 527-537.

- [15] Fratantonio, D., Rojas-Cardenas, M., Mohand, H. S. H., Barrot, C., Baldas, L., & Colin, S. (2018). Molecular tagging velocimetry for confined rarefied gas flows: Phosphorescence emission measurements at low pressure. *Experimental Thermal and Fluid Science*, 99, 510-524.
- [16] Marrero, T. R., Mason, E. A. J. J. o. P., & Data, C. R. (1972). *Gaseous diffusion coefficients*. 1(1), 3-118.
- [17] Hu, H., Zhu, J., Yang, F., Chen, Z., Deng, M., Weng, L., . . . Zhou, Y. (2019). A robust etb-type metal–organic framework showing polarity-exclusive adsorption of acetone over methanol for their azeotropic mixture. *Chemical Communications*, 55(46), 6495-6498. doi:10.1039/C9CC02439E
- [18] Nadykto, A. B., & Yu, F. (2003). Uptake of neutral polar vapor molecules by charged clusters/particles: Enhancement due to dipole - charge interaction. *Journal of Geophysical Research: Atmospheres*, 108(D23).
- [19] Bowen, T. C., Noble, R. D., & Falconer, J. L. J. J. o. M. S. (2004). Fundamentals and applications of pervaporation through zeolite membranes. 245(1-2), 1-33.
- [20] Almy, G., & Anderson, S. (1940). Lifetime of fluorescence in diacetyl and acetone. *The Journal of Chemical Physics*, 8(10), 805-814.
- [21] Mirzaei, M., Dam, N., & van de Water, W. J. P. R. E. (2012). Molecular tagging velocimetry in turbulence using biacetyl. 86(4), 046318.
- [22] Fratantonio, D., Yeachana, V., Rojas-Cárdenas, M., Colin, S., & Barrot-Lattes, C. (2018). Imaging technique for measuring the diffusion of diacetyl and acetone vapors at low pressures. Paper presented at the 31st International Symposium on Rarefied Gas Dynamics, Glasgow, United Kingdom. <https://hal.archives-ouvertes.fr/hal-03517063>
- [23] Fratantonio, D. (2019). Molecular tagging velocimetry in rarefied and confined gas flows. Ph.D. Thesis, Université de Toulouse, Toulouse, France.