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► **To cite this version:**

Dominique Fratantonio, Stefan K Stefanov, Marcos Rojas-Cárdenas, Christine Barrot-Lattes, Stéphane Colin. Photoluminescence models in direct simulation Monte Carlo for molecular tagging techniques. 5th European Conference on Microfluidics (μ Flu18) and 3rd European Conference on Non-Equilibrium Gas Flows (NEGF18), Feb 2018, Strasbourg, France. hal-02062537

HAL Id: hal-02062537

<https://hal.science/hal-02062537>

Submitted on 8 Mar 2019

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PHOTOLUMINESCENT MODELS IN DIRECT SIMULATIONS MONTE CARLO FOR MOLECULAR TAGGING TECHNIQUES

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KEY WORDS

Molecular tagging, photoluminescence, Direct Simulation Monte Carlo.

SHORT SUMMARY

Molecular tagging techniques are promising tools for providing local measurements of pressure, temperature and velocity fields in rarefied gas flows. These techniques are based on the optical collection of photons emitted by a molecular tracer, such as acetone or diacetyl vapor, when excited by UV light. The measurement of the gas properties of interest is carried out by exploiting the intensity, the lifetime and/or the spatial displacement of the emitting tracer. A key point for the success of these techniques resides on the ability of accurately extracting the information from the distribution of the emitted light. In this work, a simple statistical model for describing the photoluminescence process is proposed and implemented in the framework of the Direct Simulation Monte Carlo method. New experimental data on acetone and diacetyl emission are compared to the numerical simulations.

EXTENDED ABSTRACT

Introduction

The subject of rarefied gas flows is of central interest in the field of microfluidics. Especially in the last decades, the development of microfabrication techniques increased the number of practical applications and pushed the interest of the scientific community in further analysing the gas behaviour at the micro-scale. Microdevices that deal with micro-gas flows already exist, such as Knudsen pumps [1], micro-actuators for aeronautical and space applications [2] and micro heat exchangers [3].

By shrinking the dimensions of the system, the degree of rarefaction increases as a consequence of a reduction of the number of intermolecular collisions. When the mean free path λ , namely the average distance travelled by a molecule between two consecutive collisions, is comparable or higher than the characteristic dimension of the system L_c , thermodynamic non-equilibrium phenomena appear, thus heat and mass transfers in the system cannot be modelled by using the classical continuum assumption. Since many micro-devices are constituted by elements that deal with gas-wall interactions, such as micro-channels, understanding how the interaction between gas molecules and solid surface changes the macroscopic properties of the flow as the level of rarefaction increases is of fundamental importance. Indeed, local thermodynamic disequilibria at the wall produce a velocity slip and a temperature jump [4].

Theoretical models for describing the effect of local thermodynamic non-equilibrium have been developed since more than one century [5]. However, these mathematical models need to be validated and adjusted by means of experimental data. Indeed, most of the experimental studies available in literature related to rarefied gas flows in micro-channels provide measurement of global quantities, such as mass flow rates, inlet and outlet pressure and temperature. Therefore, the effect of the local thermodynamic disequilibrium is only indirectly evaluated [6, 7]. To the best of our knowledge, there are no direct measurements of velocity profiles in confined rarefied gas flows.

In this context, the molecular tagging technique is a promising tool for answering to this urgent need of local experimental data on confined rarefied gas flows. It is a low-intrusive, optical technique based on the exploitation of a molecular tracer that is able to emit light in the visible spectrum when excited by a UV laser source. Common molecular tracers employed are acetone and diacetyl vapor [8, 9]. By following the displacement of the tagged tracer molecules, the molecular tagging velocimetry (MTV) could provide direct measurement of the slip velocity at the wall, as well as the overall velocity profile in micro-channels. The molecular tagging thermometry (MTT) can instead provide local information on the gas temperature distribution and on temperature jump at the wall by measuring the intensity and the duration of the light emitted by the tracer. The MTV has already been applied to liquid flows and non-rarefied gas flows in channels [10, 11] while the MTT has seen applications in liquid flows and gas flows in combustion chambers [12, 13]. No applications on rarefied gas flows in confined systems exist at this moment, to the extent of our knowledge. However, recent experimental studies provided evidences on the feasibility of the application of the MTV and MTT to gas flows in rarefied conditions [14, 15].

A key point for the success of these techniques resides on the accurate reconstruction of the velocity and temperature distribution of the gas from the light emission of tracer. Strictly speaking, the MTV and MTT provide information about the intensity, the lifetime and the spatial distribution of the tracer emission, but does not provide a direct measurement of the fluid dynamic and thermodynamic properties of the gas flow under analysis. For instance, the molecular tagging velocimetry (MTV) requires an advection-diffusion modelling of the tracer displacement for reconstructing the velocity field from the tracer displacement field [16]. However, this methodology of post-processing the data needs an accurate estimation of the diffusion coefficient and the intramolecular processes that occur in the tracer molecules can affect the diffusion mechanisms, e.g., through possible variations of temperature or even molecular dissociation produced by the photon excitation. For the molecular tagging thermometry (MTT), the gas temperature measurement is based on the relationship between the tracer temperature and the intensity or the lifetime of the tracer emission. However, the photoluminescence process is characterized by intramolecular transitions that can bias the tracer temperature from the gas temperature and could possibly modify the gas temperature itself in a significant manner. For these reasons, designing a mathematical model that takes into account for the most important intramolecular processes is necessary to exploit the experimental data on the tracer emission for reconstructing the gas properties of interest.

In this work, new experimental data on acetone and diacetyl emission are presented and a novel implementation of photoluminescence models in the framework of the Direct Simulation Monte Carlo (DSMC) method is proposed. DSMC simulations are carried out for a mixture of gas and tracer molecules. The intermolecular collisions and the intramolecular transitions are both simulated for better understanding how these two phenomena interact and affect the pressure and temperature distribution of the gas-tracer mixture, the molecular diffusion of the tracer, and the spatial and temporal evolution of the tracer emission as well. Various events characterize the photoluminescence phenomenon. Starting from the photon absorption, the molecular excitation produces a change in the electronic state as well as an activation of rotational and vibrational modes. Following the excitation, many events can occur: intramolecular transitions, e.g., vibrational relaxations and intersystem crossing transitions, non-radiative de-excitations, e.g., molecular quenching, and radiative de-excitations, such as fluorescence and phosphorescence emissions [17]. A mathematical representation that describes all the intramolecular processes is a difficult task. Therefore, a first basic representation that takes into account for only the excitation and the radiative emission events is here proposed. Later, the complexity of the model will be gradually increased with the aim to numerically reproduce the experimental data.

The laser excitation process occurs in a time interval of the order of a few nanoseconds. Therefore, the photon absorption happens practically instantly when compared to the de-excitation relaxation time and to the fluid dynamic characteristic times, such as heat transfers and molecular diffusion. In the DSMC representation, each model particle corresponds to a group of molecules. Following the photon absorption, only a percentage of molecules belonging to a particle are excited.

The modelling of the radiative de-excitation event is based on experimental observations. In literature, experimental studies demonstrated that the luminescence time evolution given by acetone and diacetyl molecules fits an exponential or bi-exponential function [18, 19]. The experimental data presented in this work confirm this fact. From these observations, the emission rate $I(t)$ of photons provided by one model particle is hypothesized to be proportional to the number of excited molecules $n_{ex}(t)$ contained in it, with the constant of proportionality corresponding to the emission rate constant k [20]. Starting from this hypothesis, a probability density function (PDF) that defines the number of occurrence n of radiative de-excitations is associated to each model particle of the DSMC simulation. In order to reproduce the exponential decay of the excited molecule population that is experimentally observed, a Poisson probability distribution is chosen:

$$P_{Poisson}(n) = \frac{\mu(t)^n}{n!} e^{-\mu(t)}, \quad (1)$$

where μ is the average number of occurrence in a time interval Δt , such that $\mu = I(t)\Delta t = kn_{ex}(t)\Delta t$. The acceptance-rejection method [21] is then applied to generate the random variable n . As a result of this choice, the numerical simulation provides an exponential evolution of the light emission with a characteristic time equals to $1/k$. Figure 1 provides an example of experimental result and DSMC simulation of the luminescence emission of diacetyl excited by a laser beam. The comparison reveals that the experimental data are characterized by a higher dispersion of light with respect to the DSMC result. This is supposed to be due to noise produced by background light emission of diacetyl molecules absorbed by the walls of the experimental cell. This source of noise is currently not modelled in the DSMC simulation, and it will be considered in future developments.

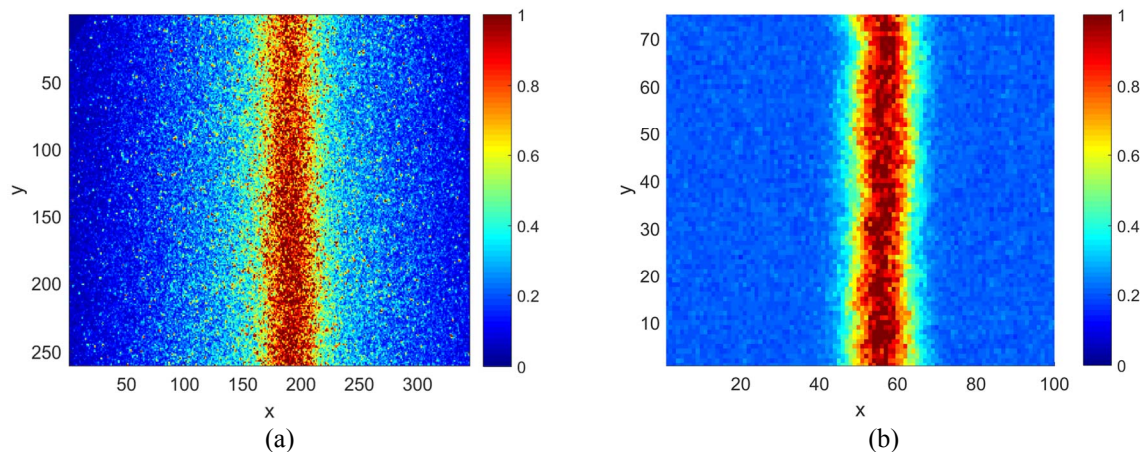


Figure 1: (a) experimental data and (b) DSMC simulation on the light emission of diacetyl vapor at pressure $p = 1000$ Pa and $50 \mu s$ after the laser beam excitation.

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. The second author, S.S. acknowledges the partial support of the Bulgarian NSF under Grant No. DN 02/7-2016.

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