

# Aerosol assisted plasma deposition: is the process controlled by the misty plasma physics?

L. Pontvianne<sup>1,2,3</sup>, P-A. Goutal<sup>1</sup>, L. Valdovinos Aguilar<sup>1</sup>, M.L. Kahn<sup>4</sup>, M. Cavarroc<sup>3</sup>, L. Stafford<sup>2</sup>, R. Clergereaux<sup>1</sup>

<sup>1</sup>Laboratoire Plasma et Conversion d'Energie, Laplace, Université Paul Sabatier, Toulouse, France

<sup>2</sup>Département de Physique, Université de Montréal, Montréal, Québec, Canada

<sup>3</sup>Safran Tech, Magny-Les-Hameaux, France

<sup>4</sup>Laboratoire de Chimie de Coordination, LCC, CNRS, Toulouse, France

**Abstract:** A lot of recent plasma deposition processes involve the injection of liquid aerosols, i.e. of droplets containing the thin film precursor. This process can be described as a misty plasma. It involves notably droplet charging, electrostatic confinement in the plasma bulk and their ballistic transport. Here, we will discuss how the process is controlled by the electrostatic sheath in a RF capacitive low-pressure plasma.

## 1. Introduction

Plasma thin film deposition is an interesting way of obtaining various coatings [1]. For example, PE-CVD enables to form various organic, organosilicon or silica-like thin films with a high degree of control. However, it needs to use gases or vapors of liquids and it is difficult to inject liquid mixtures, solutions or colloids of paramount for multifunctional surfaces. A new approach consists in a pulsed injection of an aerosol of liquid droplets in a low pressure plasma reactor [2].

Direct Liquid Injection (DLI) enables to inject in a pulsed mode liquid independently of its composition, physico-chemical properties and stability.

An isolated droplet can be submitted to a lot of mechanisms from the droplet charging to its electrostatic confinement [3]. Depending on these mechanisms, the thin film deposition can be heavily affected. To better understand the plasma-droplet interactions, pentane aerosols are introduced in a pulsed mode into a low-pressure RF capacitive plasma. The structure of the Diamond-Like-Carbon (DLC) coatings varies with the working pressure produced with a continuous injection of argon.

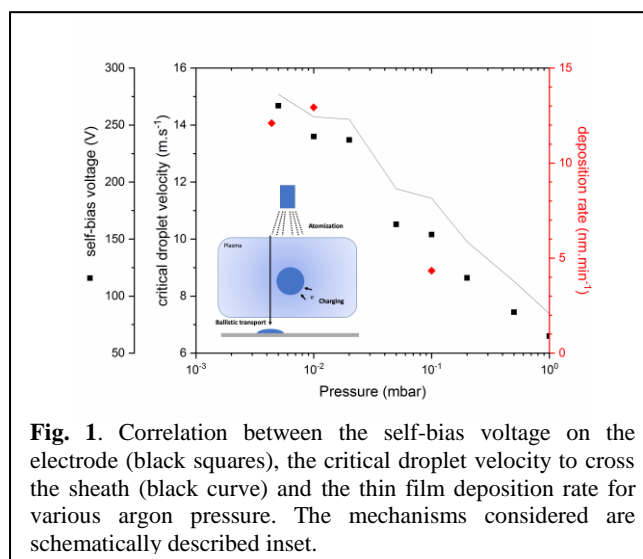
## 2. Methods

The set-up used in this study was previously described [3]. It consists in an asymmetric capacitively-coupled RadioFrequency (RF, 13.56 MHz) plasma reactor coupled with a pulsed DLI system. It is ignited at a working pressure ranging from  $5 \cdot 10^{-3}$  to  $1 \cdot 10^{-1}$  mbar, pressure fixed by a continuous injection of argon, and at 100 W with the matching box in automatic mode.

Coatings are collected onto silicon substrates and analyzed by profilometry, optical microscopy and FTIR.

## 3. Results and Discussion

With the injection of droplets from the aerosol into the plasma, many interactions occur. The coating deposition rates were determined for different argon pressures. With the increase in pressure from  $1 \cdot 10^{-2}$  to  $1 \cdot 10^{-1}$  mbar, the deposition rate drops from 12 to 4 nm.min<sup>-1</sup>. This behavior is attributed to plasma-droplet interactions. Indeed, as schematized inset Figure 1, charged droplets can be confined in the plasma volume or go through it, depending on the sheath repulsion. Though a simple simulation of the



**Fig. 1.** Correlation between the self-bias voltage on the electrode (black squares), the critical droplet velocity to cross the sheath (black curve) and the thin film deposition rate for various argon pressure. The mechanisms considered are schematically described inset.

balance of forces (considering that the electric field is mainly controlled by the self-bias voltage on the electrode) always show a predominance of the electrostatic repulsion. However, depending on the initial velocity of the aerosol and the self-bias voltage (Figure 1), some droplets can exceed a critical droplet velocity and be ballistically transported to the substrate, drying directly on the substrate and thus limiting the deposition rate. It shows that the aerosol assisted plasma process is controlled by the sheath.

## 4. Conclusion

Misty plasma physics is controlling the aerosol assisted plasma deposition. Depending on the film composition, one would expect an efficient confinement of the droplets in the plasma volume and thus the liquid evaporation or their direct transport to the substrate.

## Acknowledgement

This work was financially supported by the physics department of Université de Montréal and Safran.

## References

- [1] P. Nguyen-Tri et al., Int. J. Corros., **2018**, 1-19 (2018)
- [2] G. Carnide et al., Coatings (2023)
- [3] G. Carnide et al., Plas. Chem. And Pas. Processes, **44**, 1343-1356 (2024)