OSCILLATING MOVEMENT IN CH₄-N₂ DUSTY RF PLASMAS

J.-F. LAGRANGE^{1*}, I. GERAUD-GRENIER¹, F. FAUBERT², V. MASSEREAU-GUILBAUD¹

¹GREMI, Groupe de Recherche sur l'Energétique des Milieux Ionisés, UMR 7344, CNRS/Université d'Orléans, Site de Bourges, Faculté des Sciences, Rue Gaston Berger, BP 4043, 18028 Bourges Cedex, France ²IUT de Bourges, Département Mesures Physiques, 63 avenue de Lattre de Tassigny,

18020 Bourges Cedex, France

*e-mail: jean-francois.lagrange@univ-orleans.fr

ABSTRACT

Dust particles produced by Physical Enhanced Chemical Vapour Deposition (PECVD) in a capacitive discharge (13.56 MHz), can show a particular behaviour for specific discharge conditions and gas composition. Therefore, at a pressure of 120 Pa in a CH_4-N_2 mixture injected with a constant total gas flow rate of 5.6 sccm and a RF power of 80 W, the particles located above the grounded electrode have a periodical and vertical movement throughout the plasma bulk. This phenomenon only occurs for the following geometry of the grounded electrode: presence of through holes in the plate (used for particle collect) put on the grounded electrode.

The movement is linked to the competition between the forces acting on particles. Holes work as attractive poles and disturb the "classical" competition of forces, leading to the particle levitation in the cloud near the grounded electrode. We will discuss about the effect of the holes presence making the cloud above the grounded electrode moves vertically.

1. INTRODUCTION

Dusty plasmas were firstly studied in glow discharges [1]. In 1980 years, it was put in evidence as a problem, in silane plasmas etching and deposition processes for microelectronic applications [2,3]. Researches on dusty plasmas are investigated for various applications such as physico-chemistry and astrophysics [4]. Dusty plasmas can be used for particle production which is incorporated in materials *e.g.* incorporation of gold or silver nanoparticles on silicon for antireflection nanostructure in solar cells [5].

Behaviour and growth mechanisms of particles have to be understood in order to optimise processes. This optimisation is also necessary for a better comprehension of instability mechanisms occurring in dusty plasmas.

This work is an experimental approach of oscillating phenomena of particles, which is a kind of instabilities, and follows a previous study of this phenomenon [6,7]. Particles are produced by PECVD in our vacuum chamber in low pressure of CH₄-N₂ (30% of methane). Under force actions, particles are trapped at the boundary sheath under the upper electrode (RF electrode) and also at the boundary sheath above the lower electrode (grounded electrode). The particles in the cloud above the grounded oscillating and electrode show vertical movements that make them go upward to the upper cloud (under the RF electrode). The movement of the particles is filmed, in order to follow the position evolution of the particle cloud front. Then, it is correlated with electrical parameters in order to identify the origins of the lower cloud movement. The effect of the holes in the particle movement is studied.

2. EXPERIMENTAL SETUP

Particles are generated in a cylindrical vacuum chamber (40 cm in diameter and 40 cm in height) in a RF (13.56 MHz) discharge. The incident RF power is 80 W, the pressure 120 Pa and the gas flow rate of the gas mixture containing 30% of methane is 5.6 sccm. The capacitive coupling with the RF generator and the asymmetry of the electrodes induce a negative DC self-bias voltage (V_{dc}) on the powered electrode. The temporal evolution of the V_{dc} is recorded with an oscilloscope (Tektronix DPO5034).

Particle presence is put in evidence by laser light scattering (Ar⁺ laser, $\lambda = 514.5$ nm). The laser spot is expanded with a beam expander in order to produce a rectangular laser beam (3 cm in height and 0.5 cm in width). This beam is directed between electrodes and at their central vertical axis. The particle movement between electrodes is recorded with a camera (50 frames per second) perpendicularly to the laser beam.

Discharge is engaged between two parallel and plane electrodes, at 1.5 cm away from each other (Fig. 1). A plate containing holes is put on the grounded electrode for the particle collect at the end of the experiment. Different configurations have been investigated, in order to show the influence of the presence of holes in the plate on the oscillating movement of the lower cloud.



Fig. 1 Example of discharge configuration. Particle clouds appear in green. On this picture, a drilled plate is put on the grounded electrode.

The three plate configurations on the grounded electrode are shown in Fig. 2:

- (A) configuration: a stainless steel plate with holes is put on the grounded electrode (Fig. 1). Holes are through the plate and not obstructed, except for the central one.

- (B) configuration: it's the same configuration as the previous one, but there are not through holes.

- (C) configuration: a stainless steel plate without any holes is put on the grounded electrode.



3. PARTICLE TRAPPING NEAR ELECTRODES

Decomposition of the CH_4 - N_2 mixture in the discharge produces nanometer scale to micrometer scale particles. In a low pressure plasma, electron mass is very low in comparison to the ion one, the electron mobility in the discharge is higher than the ion one and particles become negatively charged [8]. Under force action [9,10], particles levitate into two parallel clouds at the boundary sheaths (Fig. 3). The forces are:

- Gravity force F_g which works downwards, in the direction to the grounded electrode.

- Thermophoresis force $F_{\rm th}$ which pushes particles to cold parts of the discharge and the vacuum chamber. In our vacuum chamber, it works downwards.

- Electrostatic force due to the repulsive effect between particles.

- Electric force F_e due to the potential difference between sheaths which pushes particles to the plasma bulk.

- Ion drag force F_i due to the positive ions attracted by the negative DC self-bias voltage and the ground potential. It pushes the particles toward the electrodes.

- Drag force generated by neutral gas flow. In our condition, the gas flow is very low, so this force can be neglected.





4. RESULTS AND DISCUSSIONS

Figure 4 shows the particle clouds in the different "(A)(B)(C)" configurations described in Fig. 2. The presence of holes in the plate put on the grounded electrode modifies the particle cloud shape (Fig.4. A and B). Particles of the lower cloud enter in the holes leading to the formation of arches. The arches are much more accentuated in the (A) case. Moreover, the lower cloud in (B) case looks more uniform. In Fig. 4.

C, particle repartition is uniform in the clouds and parallel to the electrodes.



Fig. 4 Pictures of the discharge for (A) configuration and (B) configuration. The pictures are in grey scale and particles clouds appear in light grey and white.

(A) configuration

The oscillating and vertical movement of the lower cloud occurs only in the (A) configuration: the lower cloud periodically goes up to the upper cloud and then goes back above the grounded electrode.

In a previous study [6] carried out in (A) configuration, we show the influence of the force competition, which involves the vertical movement of the lower particle cloud. We made the hypothesis that the attraction of the lower cloud by the upper one is mainly guided by an electrostatic attraction or by plasma modifications. In this experimental condition, the attraction is coupled with a too weak ion drag force to maintain the lower particle cloud at the grounded electrode sheath.

Particles are negatively charged by the trapping of plasma bulk electrons. Their formation and movement in the discharge leads to electrical modifications such as V_{dc} values. Moreover, the recording of the oscillations by the camera allows to get the positions of the lower cloud front between the electrodes. The V_{dc} values and

the upper cloud positions from the grounded electrode (GND electrode) are reported in Fig. 5 for some oscillations.



Fig. 5 Time evolution of V_{dc} and position of the lower cloud front during oscillating movement

This V_{dc} variation is almost sinusoidal with a frequency of 0.58 Hz. Lower values correspond to the position of the cloud above the grounded electrode, and upper values correspond to the position of the cloud under the RF electrode. The movement occurs along the following steps:

- a few particles leave the upper cloud and a weak and slow increase of the upper cloud thickness is observed.

- the particles located in the front of the lower cloud are submitted to the ascending ion drag force leading to the cloud rising. Particles trap electrons and $V_{\rm dc}$ decreases.

- then, particles are submitted to the downward thermophoretic force. They go back above the grounded electrode and they are again attracted by the holes.

(B) and (C) configurations

In the (B) and (C) configurations, no vertical movement of the lower cloud occurs towards the RF electrode. Different experiments have been carried out by increasing the RF power until 130 W and by modifying the distance between the electrodes.

As shown in Fig. 4 A, the presence of through holes allows the plasma to go freely under the plate (Fig. 1), which is not possible for the (B) and (C) configurations. Holes attract both particles and plasma. They act as a potential well that seems to be stronger in (A) configuration case than in the (B) one. Looking at the lower cloud, the arches are enhanced in (A) case. It can be explained by a stronger electrical field in the holes leading to a stronger stress on the cloud. Some cloud particles seem to be aspirated by the holes. The arches are much more luminous and the distance between the arch tops and the upper cloud is lower. Moreover the plasma is freely expanded under the plate leading to a lost of the power between the electrodes. That would mean, for the same incident RF power, a weaker downward ion drag force in comparison with (B) and (C) configurations. Consequently, the lower cloud position is slightly higher in the plasma. When the cloud thickens, the particles located in the cloud front are submitted to the ascendant ion drag force. The lower cloud front is attracted toward the RF electrode and all the cloud particles are dragged. When the lower cloud goes up in the discharge, it escapes the hole attraction. Then. this cloud goes downward bv thermophoresis force and hole attraction.

5. CONCLUSION

The attractive role of the holes has been pointed out. For an oscillating movement, it is necessary to have particles going throughout the plate holes, and to have an expansion of plasma under the plate. Thus, power is lost in plasma expansion which lowers the power between the electrodes. In that case, the distance between the arch tops of the lower cloud and the upper cloud is smaller (in comparison with (B) configuration at a same RF power). So, ion drag force is weaker and it is the triggering factor which can help lower cloud to get out of holes attraction.

6. ACKNOLEDGMENT

This work is supported by the Agence Nationale de la Recherche, INDIGO project (ANR-11-JS09-010-01).

REFERENCES

[1] I. Langmuir, C. G. Found, and A. F. Dittmer, "a new type of electric discharge: the streamer discharge", Sci, **60**, 392-4, 1924.

[2] G. S. Selwyn and J. Singh, "Plasma-enhanced photoemission in argon discharges: Signal characterization and silicon doping effects", J. Vac. Sci. Technol. A, **7**, 982-986, 1989.

[3] K. G. Spears, T. J. Robinson, and R. M. Roth, "Particle Distributions and Laser-Particle Interactions in an RF Discharge of Silane", IEEE Trans. Plasma Sci., **14**, 179-187, 1986.

[4] C. D. Pintassilgo and J. Loureiro, "Kinetic study of a N2–CH4 afterglow plasma for production of N-containing hydrocarbon species

of Titan's atmosphere", Adv. Space Res., 46, 657-671, 2010.

[5] K. Nishioka, T. Sueto, and N. Saito, "Formation of antireflection nanostructure for silicon solar cells using catalysis of single nanosized silver particle", Appl. Surf. Sci., **255**, 9504-9507, 2009.

[6] J.-F. Lagrange, O. Vallée, I. Géraud-Grenier, F. Faubert, and V. Massereau-Guilbaud, in *Etude préléminaire des oscillations spontannées dans un palsma poudreux radiofréquence (13.56 MHz) de méthane-azote*, Modélisation : Atomes, Molécules, Plasmas et Systèmes Dynamique, Bourges, 2013 (Presses Univesitaires d'Orléans).

[7] J. Pereira, V. Massereau-Guilbaud, I. Geraud-Grenier, and A. Plain, "Nitrogen effect on the dust presence and behavior in a radio frequency CH_4/N_2 discharge", J. Appl. Phys., **103**, 033301-9, 2008.

[8] Y. A. Mankelevich, M. A. Olevanov, and T. V. Rakhimova, "Dust particle coagulation mechanism in low-pressure plasma: rapid growth and saturation stage modeling", Plasma Sources Sci. Technol., **17**, 015013, 2008.

[9] I. Géraud-Grenier, V. Massereau-Guilbaud, and A. Plain, "Analysis of particulates generated in a radiofrequency methane plasma by laser light scattering and optical spectroscopy", Eur. Phys. J. App. Phys., **8**, 53-59, 1999.

[10] C. Zafiu, A. Melzer, and A. Piel, "Ion drag and thermophoretic forces acting on free falling charged particles in an rf-driven complex plasma", Phys. Plasmas, **9**, 4794-4803, 2002.