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Particle Observations and Optical Spectroscopy in Methane Radiofrequency Discharges.

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1. INTRODUCTION

The growth processes and the behavior of particles in processing plasmas have been investigated in the last few years. Particles in low pressure discharge induce contamination in microelectronic fabrication. Moreover, particles are of interest in astrophysics to study nebulae, comet tails…

In this study, the amorphous hydrogenated carbon particulates are generated in a classical planar radiofrequency (13.56 MHz) reactor in CH_4 atmosphere. Two particles populations coexist in the plasma. The first one composed of white spherical particles generate in the discharge. The second one coming from the sputtering of the coating growing onto the biased electrode are brown and look like leaves.

The time evolution of the laser beam extinction through the particulate cloud at the cathodic sheath boundary is correlated with the particulate behaviour in the discharge, with the cloud length and with the dc self bias voltage variations. The methane flow rate has a very small influence on the particulate growth and behaviour in the discharge. On the other hand, an increase of the methane pressure or of the incident power leads to a more important laser beam extinction, so to a larger particulate cloud and to a greater dc self bias variation. Moreover, the temporal evolutions of the phenomena are faster when the incident power increases.

Spectroscopic measurements have been realized. We have studied the temporal evolutions of the emission lines (CH, H_2 and H_α) in the discharge. The results have been correlated with the growth of amorphous hydrogenated carbon particulates and also with the results obtained by laser light scattering.

2. EXPERIMENTAL SETUP

The experimental set up, we have used is schematically shown in figure 1. The particles are obtained in a capacitive 13.56 MHz RF plasma generated from methane gas in a cylindrical reactor with parallel aluminium plate electrodes -2.5 cm apart-. The powered electrode, 13.5 cm in diameter acquires a dc negative self bias voltage with respect to the ground. The grounded electrode is 20 cm in diameter. In the reactor a residual total pressure lower than 10^{-3} Pa is achieved using a secondary pumping system. The methane pressure during the process is controlled by a capacitance gauge. The particle formation and optical emission lines are studied for the following experimental ranges methane pressure (20-150 Pa), methane flow rate Q (2-14 sccm) and incident rf power (40-120 W)

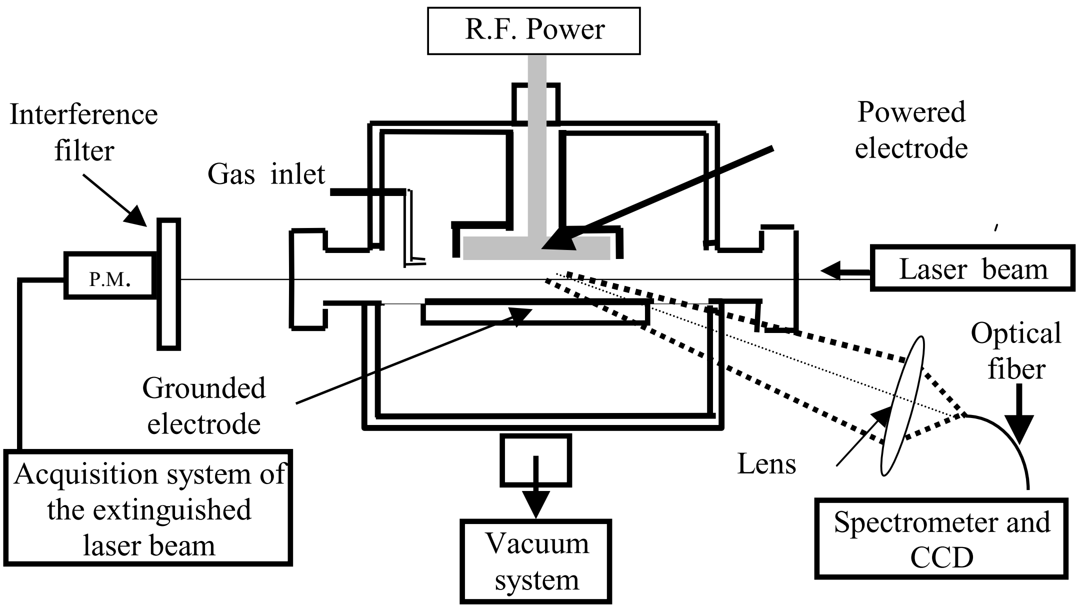

Figure 1 : Experimental set-up

The plasma optical emission is analyzed through a glass window with a lens focusing the light onto a quartz optical fiber connected to a spectrometer and coupled with an optical multichannel analyzer (OMA). During the experiment an Ar^+ laser beam (• = 514.5nm and P.=50mW) is directed through the plasma and the scattered light signal indicates the presence of particles. The laser beam extinction through the discharge is analyzed using an optical system (optical attenuator, interference filter, low noise photomultiplier) coupled to an acquisition system.

3. RESULTS AND DISCUSSION

3.1. Particle behaviour

During the experiment, two kinds of particles are generated. The first family is composed of particles generated in the plasma bulk. They are white, spherical and 0.5-1.1 µm in diameter. The second ones result from the flaking of the coating growing on the RF biased electrode. They look like brown leaves and they are 100 µm long and 10 µm wide. These brown particles fall across the discharge on the grounded electrode. The white particles have been evidenced by laser light scattering. They are trapped in clouds in the plasma bulk at the boundaries with the cathodic and anodic sheaths. To understand their behaviour in the discharge, the laser beam extinction has been studied. Particulates are mainly observed at the sheath boundaries where they are trapped in clouds resulting from the action of different forces.

The particulate appearance time at the cathodic sheath boundary is very sensitive to the experimental conditions:

- the higher the pressure and the rf incident power, the shorter the appearance time,
- the higher the methane flow rate, the longer the appearance time.

We have shown the correlation between the particle appearance times t :

- in inverse proportion to of the CH4 molecules residence times,
- with the CH4 pressure in the reactor,

The time evolution of the laser beam extinction through the particulate cloud at the cathodic sheath boundary is correlated with the particulate behaviour in the discharge, with the cloud length and with the dc self bias voltage variations. The methane flow rate has a very small influence on the particulate growth and behaviour in the discharge. On the other hand, an increase of the methane pressure or of the incident R.F. power leads to a more important laser beam extinction, so to a larger particulate cloud and to a greater dc self bias variation. Moreover, the temporal evolution of the phenomena are faster when the R.F. incident power increases.

3.2 Spectroscopic results

The temporal behaviour of the different emission lines in the 380-800nm spectral range have been analyzed as a in function of the R.F. power (40-120W) and the pressure (65-150Pa):

-.the Balmer atomic hydrogen emission lines: H_α at 656.3nm,H_β at 486.1nm and H_γ at 434.0nm.

- the Fulcher α ($d^3\ \Pi_u$-$X^3\ \Sigma_g$) molecular hydrogen band in the 590-640nm region.
- the ($G^1\ \Sigma_g^+$ -$B^1\Sigma_u^+$) molecular hydrogen band at 463.4nm.
- the C_2 Swan system ($d^3\Pi_g$ - $a^3\Pi_u$) with the Q(v'=0,v''=0) band at 516.5nm and the Q(v'=1,v''=1) at 512.9nm.
- the CH ($A^2\Delta$;v'=0 $X^2\Delta$;v''=0) system at 431.2nm .

3.2.1 Influence of the rf power.

The line intensity is increasing with the R.F. power resulting from the increase in the dissociation rate of the methane (pressure of 133Pa). But we have noticed the intensities of the different lines have different temporal time evolutions. The hydrogen line intensities continuously increase with time. But the ratio of H_2/ H_α is rapidly increasing in the range 0-120s and tends to a saturation value. The CH line intensity also increases with R.F. power but it decreases with time to a saturation value. The time evolution intensities are the consequence of many different phenomena. The discharge configuration as the electrical characteristics are modified as soon as the discharge is switched on, due to the formation and particle behaviour in the discharge, and the insulating of the electrodes.

3.2.2 Influence of the methane pressure.

The temporal evolution of the spectral line intensity has been analysed in the range 65-150Pa with an rf incident power of 80W. We have observed that the intensity of the different lines are decreasing as the pressure is increasing and, the intensities of the different lines have different time evolutions with similar behaviour as in the case of the power study. The pressure increase leads to a decrease in the electron temperature and electron number density as an increase in the concentration of the reactant species. This assumption corresponds to the fact that the particles appearance time is decreasing as the pressure is increasing.

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