Formation and transport of dust particles in ECR acetylene plasmas

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In this work, the formation and transport of carbonaceous dust particles are studied in acetylene ECR plasma using fast imaging of dust particles together with ex-situ analysis of particles samples collected during experiments. It seems that both volume and surface mechanisms are involved in particles formation. Moreover, the dynamics of dust particles exhibits particular trends and strongly influences the physical mechanisms arising in particles formation.

1. Introduction

The formation and transport of dust particles in plasmas have been intensively investigated in different fields. Since the beginning of the 90's [1] and the observation of charged dust particles in the rings of Saturn (data collected with the probes Voyager 1 and 2), astrophysists described a key role of dust particles in the interstellar media, as well as in planetary environment [2]. In 1989, Gary Selwyn explained the unexpected occurrence of dust particles in the microelectronic manufacturing industry [3]: it is spontaneously formed within the plasma chamber through chemical reactions. Numerous studies were thus performed to avoid contamination of silicon wafers during microprocessor manufacturing. More recently, dust occurrence in plasmas has gained interest regarding plasma-surface interactions in fusion devices where dust can induce several drawbacks such as tritium retention, radioactive contamination of the chamber walls or safety issues in case of vacuum or water leaks [4]. Nowadays, dusty plasmas are considered as a powerful tool to generate nanomaterials. It offers a very convenient way to generate controlled nanoparticles inclusions in an amorphous matrix such as crystalline silica nanoparticles [3] or silver nanoparticles [4] intended for photovoltaic or antibacterial applications.

However, most of the aforementioned studies were performed in moderate to high-pressure environments (capacitive discharges, dielectric barrier discharges, arc discharges, etc.). In these pressure regimes, the probability of recombination in the plasma volume is high enough to sustain rapid particle growth through chemical paths. Despite these considerations, dust particle occurrence in acetylene low-pressure electron cyclotron resonance (ECR) plasmas has been observed [5], thus asking new questions on dust formation and transport in plasmas.

In this work, fast imaging of dust particles and ex-situ analyses were performed in acetylene ECR plasma. Fast imaging revealed a noticeable particle transport along the plasma chamber that allows describing the main forces on dust particles in such plasma. Moreover, the obtained results indicate that dust particles formation arises first from volume (growth) and secondly from surfaces (remobilization). Dust particles transport along the plasma chamber plays an important role in this second mechanism itself, which indicates that formation and transport are in close correlation with each other.

2. Experimental Setup

Experiments are performed in the reactor illustrated in figure 1 consisting of a stainless steel vacuum chamber with inner dimensions of $110\times16\times12$ cm^3 along the X, Y and Z-axes, respectively. The gas inlet and outlet are located on both sides of the XZ planes. Gas injection is set by mass flow controllers and the pumping system is based on a turbomolecular and rotary pump combination. For the present work, the acetylene flow rate was set to 1.2 sccm resulting in a working pressure of 0.6 mTorr. The microwave power, tuned between 125 and 200 W, is injected from the X=0, Y=0 and Z=0 along a 110 cm long antenna. The latter is parallel to a samarium-cobalt racetrack magnet. Plasma is excited in the electron cyclotron resonance (ECR) region (B=875 Gauss) located a few cm above the antenna (f_{ex}=2.45 GHz).

A glass window covers a large part of the top of the reactor, which allows analyzing the incandescent particles dynamics. The latter is done through fast video imaging using a Photron SA.5 camera set parallel to the Z axis equipped with a Nikkor 55mm

f2.8 lens. In the present experiment, there is no need to illuminate the particles with an external light source such as a laser sheet. Indeed, dust particles are incandescent and thus emit their own light. This phenomenon is not yet fully understood and currently under investigations. Movies were acquired at a rate of 20.000 fps for a corresponding observation area of few cm^2.
For ex-situ analyses, throughout the plasma discharge, stainless steel substrates were placed at different locations in the bottom of the reactor to collect and then analyze dust particles with ex situ electron microscopy (SEM and TEM).

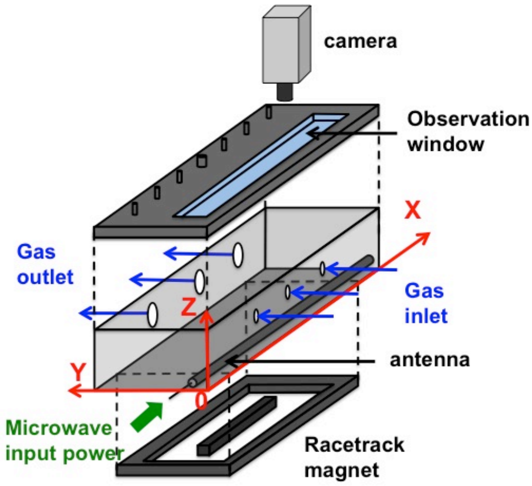
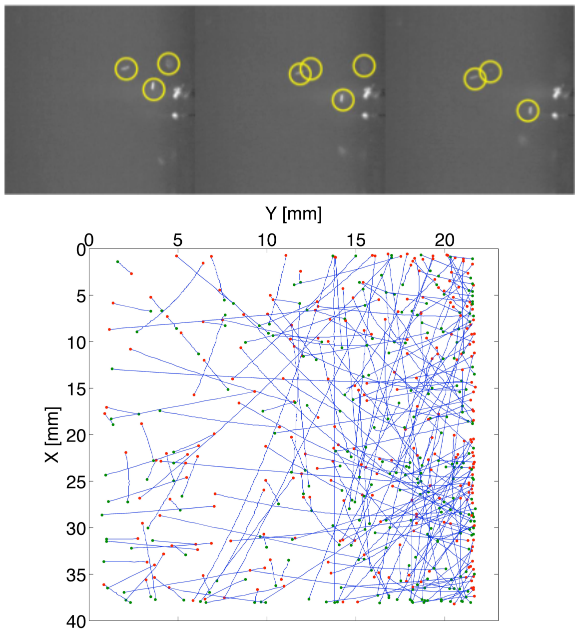


Figure 1: Experimental setup for the fast camera transport analyses.

3. Dust particle dynamics

Screenshots of recorded movies are reported in figure 2A. They are analyzed by the TRACE algorithm [6], which automatically recovers the whole dust particles trajectories and thus permits the calculation of velocity and acceleration distributions. Following this method, the large number of analyzed trajectories (several hundreds) ensures statistically relevant results.
Most of the particles were found to be located in a focal X-Y plane of the camera at Z=3 cm from the bottom of the reactor. A typical example of a superimposition of recovered particles trajectories is depicted in figure 2B For this experiment, the movie duration was about 0.66 s (13175 frames) with the camera fixed at the position X=392 mm.

A)



B)

Figure 2: A) fast camera images of dust particles in the ECR plasma. B) Recovered particle trajectories are obtained using the TRACE algorithm.

Most of the trajectories are rectilinear. It suggests that the magnetic field has not a direct key role in the transport of dust particles. Moreover, at the position X=392 mm, as shown in figure 3, the velocity distributions of dust particles along X and Y show strong discrepancies. While it is roughly centered on zero for the Y component, the distribution is clearly shifted towards negative values for the X component. It can be confirmed by comparing the average and median values for both distributions, respectively -0.1 $m.s^{-1}$ and -0.5 $m.s^{-1}$ along the Y-axis and -3.6 $m.s^{-1}$ and -3.7 $m.s^{-1}$ along the X-axis. These results clearly show a preferential transport of particles towards decreasing X values.

Estimation of the particles acceleration is of primary importance to investigate their dynamics since it provides information regarding the forces acting on the particles. Acceleration distributions depicted in figure 4, present once again a different behavior between the X and Y directions. Firstly, almost every dust particle is subjected to a negative acceleration along X whereas along Y, the distribution is centered on zero. Moreover, the magnitude of the acceleration is noticeably greater along the X-axis. It indicates that at least one force is responsible of the particle acceleration in the longitudinal direction.

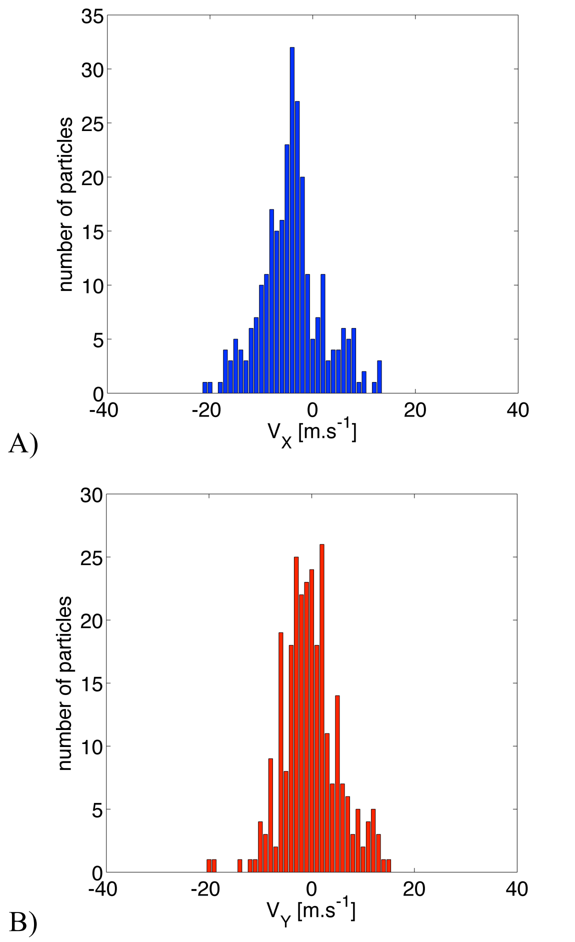


Figure 3: Velocity distributions along A) X and B) Y.

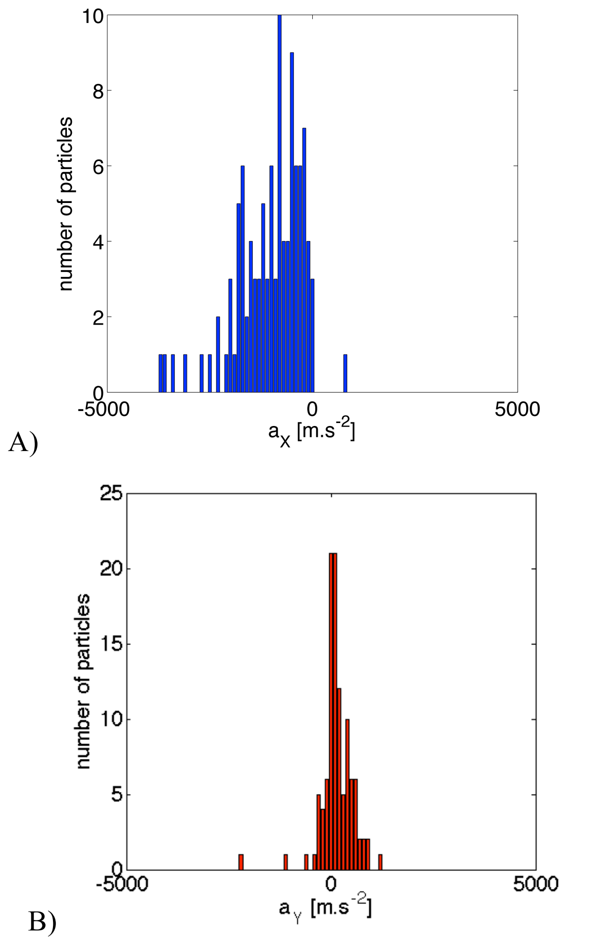


Figure 4: Acceleration distributions along A) X and B) Y.

Recent Langmuir probe measurements in argon performed at different locations along X revealed the presence of a small electric field related to a gradient in the plasma potential. Its value was estimated to be around few volts per meter. Considering this field and the median value of the acceleration (-1000 m.s^{-2}), one can estimated the size of the accelerated particles to be around 10 nm for graphitic (ρ=2000 kg.m^{-3}), spherical, and negatively charged (47 electrons considering OML model in a T_e=3 eV plasma) particles. It could explain the observed particles acceleration through the well-known electrical force.

4. Ex-situ analysis of collected samples

Figure 6 shows SEM micrographs of typical carbon clusters collected in the edge of the magnetic racetrack. One can identify spherical dust particles with an average radius of 10 or 50 nm (figure 6A and B) in good agreement with the previous size estimations. Moreover, spherical particles with a diameter of about 1 µm were also observed (figure 6C).

These three kinds of dust particles look like those formed by the classical growth model of plasma volume recombination in RF capacitively coupled discharges, i.e. nucleation, aggregation, coalescence and surface deposition [7].

On the other hand, figure 6D presents a filament-like layered aggregate of a few tens of µm in length. Such geometrical configuration probably originates from a different – and possibly more complex – formation mechanism. Indeed, visual observations during experiments also showed that a large amount of incandescent carbonaceous materials are deposited in the edge of the magnetic field and, more accurately, at a specific location on the bottom of the reactor to form a so-called “hot spot” (HS). A picture of the latter is presented in figure 5. Further investigations using fast imaging revealed that this location is a favored place for particle deposition and remobilization into the plasma volume. Note that such mechanism shows similarities with erosion/re-deposition mechanism involved during plasma-wall interaction in fusion devices [2].

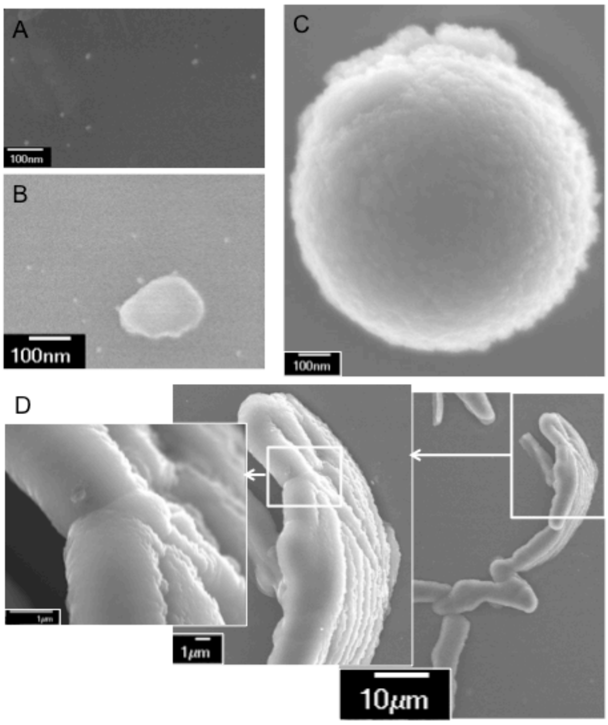


Figure 6: SEM pictures of typical collected carbon clusters such as a spherical dust particle (A and B) and carbon layered filament (C and insets).

One could hypothesize that carbonaceous clusters aggregate at the HS into long filaments and are then subjected to constraints from a harsh environment (etching from charge particles fluxes, resistive heating). Subsequently, matter is ejected within the plasma volume and may participate in new hydrocarbon cluster formation. These observations indicate that the observed moving particles can be of different nature and size and that several physical mechanisms are involved in their formation.

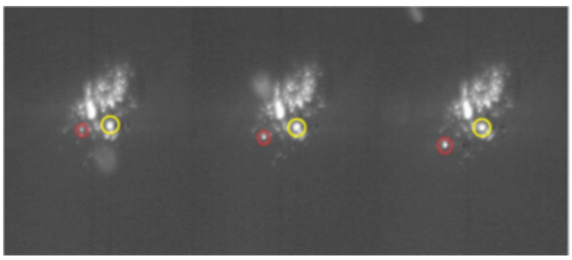


Figure 5: Fast camera images of the HS where surface deposition diffusion (yellow) and remobilization (red) of carbon clusters are observed.

5. Conclusion

Investigations of dust particles formation and transport in acetylene ECR plasma have been carried out. It has been shown that incandescent particles move with high velocities in a preferential direction due to at least one force, supposedly the electrical force. Particles transport also favors their deposition onto preferential locations in which remobilization of deposited matter may also take place. Then, ex-situ analyses revealed two different kinds of dust particles: spherical ones probably formed within the plasma volume, and flake-like particles, indicating that several mechanisms contribute to their formation and growth.

6. References

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