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Comparison between carbon dust produced in laboratory plasmas and in Tore Supra

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Abstract

Laboratory experiments are proposed to understand the growth mechanisms of spherical carbonaceous dust observed in Tokamaks with inside wall elements in graphite materials. The sputtering process is used to form continuous carbon vapours. Their cooling in the plasmas gives rise to carbon clusters which size goes increasing with time. In the nanometer scale range, the obtained primary particles are spherical. They can also agglomerate in the plasma likely by coulomb attraction and form spherical aggregates of higher size. A comparison between the carbon structure of these dust grains and of some dust samples collected on the toroidal pumped limiter surface of Tore Supra is also proposed. The differences are discussed.

keywords: Plasma discharge, powder formation, sputtering, carbon

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1- Introduction

Among the carbonaceous fragments usually collected in Tokamaks, one can observe the presence of spherical nanosized particulates¹. Their precursors result from multiple collisions between the reactive carbon atoms and hydrocarbon radicals present in the edge plasmas and originating from the Tokamak graphite wall physical and chemical erosion. When carbon vapours are injected in plasmas, their cooling and then, their condensation can generate different carbon arrangements: linear and/or spherical molecules like fullerene², basic structural units (BSU) composed of stacks of few aromatics layers³, onion-like structure composed of bent and nearly concentrically arranged aromatic layers³, clusters where carbon atoms are structured diamond-like⁴ (DLC) and where heteroatoms can be incorporated... When discharges are produced in hydrocarbon gases as methane, ethylene, acetylene desorbed by Tokamak graphite walls, the electron collisions lead to the formation of multiple molecule fragments and radicals. In particular, the presence of carbon triple bonding favouring

polymerisation reactions⁵ and of polycyclic aromatic hydrocarbon⁶ (PAH) are other sources of dust formation.

The aim of the presented work is to characterize the dust formation from carbon vapours developed in different conditions: laboratory and Tore Supra (TS) conditions. In our experiments, carbon is injected in argon plasmas through cathode sputtering of glow discharges. The carbon vapour condensation produces spherical primary particles (PP) in the nanometer scale range. Their agglomeration forming bigger particulates is also observed and assigned to coulomb attraction when the PP have fluctuating surface charges. The PP carbon structure is determined by XANES analysis (X-ray absorption near edge spectroscopy) as well as the carbon structure of a dust sample taken in December 2002, on the toroidal pumped limiter (TPL) surface of TS⁷. This graphite wall (TPL), actively cooled by pressurized water at 120°C undergoes high heat load during plasma operations^{8,9}. Some regions on the TPL surface can be covered by dust layers.

The observed differences between the carbon structure of the dust samples formed in glow discharges and in TS are discussed.

2- Laboratory experimental arrangements

DC glow discharges are performed between two plane electrodes of 10 cm diameter, the cathode being in graphite and the grounded anode situated at the bottom, in stainless steel. They can be separated by different distances, either $d_0 = 5$ cm corresponding to the minimum of Paschen law for the argon pressure of $P_{Ar} = 0.6$ mbar or $d_1 = 14$ cm. In the former case, the regulated discharge current is $I = 70$ mA, the adjusting graphite cathode bias being $V = -545$ V and in the latter case, $I = 80$ mA with $V = -530$ V. The electron density and temperature are respectively, in the range of: $n_e \sim 10^{16} \text{ m}^{-3}$, $T_e \sim 1-2$ eV (ionized fraction $\sim 10^{-5}$). The neutral collision mean free path is $L_{Ar-Ar} \sim 125 \text{ } \mu\text{m}$. The carbon source is provided by ions and energetic charge exchange (CX) neutral sputtering since the CX length $L_{CX} \sim 200 \text{ } \mu\text{m}$ is smaller than the sheath thickness (centimeter range). During the working time, the cathode and anode temperature measured with thermocouples, never exceeds 150°C and 80°C, respectively. The dust are collected on the anode surface. The optical spectroscopy control of C₂ Swann band, C₂ molecule being a dust precursor, allowed us to optimize the discharge parameters for the dust production in the case where $d_0 = 5$ cm.

3- Results and discussion:

a) Shape analysis of laboratory and Tore Supra dust samples

The scanning electron microscopy picture (SEM) of **Fig. 1** shows an example of PP produced after several successive 10 min glow discharges for $d_0 = 5$ cm. Their size is $\phi \sim (38 \pm 5)$ nm. If it is assumed that the PP stay in the plasma during the time discharge and that their growth is due to a homogeneous carbon deposition, one can estimate the carbon flux being $\Gamma_C \sim 10^{19} \text{ m}^{-2}\text{s}^{-1}$.

The transmission electron microscopy (TEM) picture of **Fig. 2** shows spherical aggregates of size $\Phi \sim 190$ nm, produced after several successive 10 min glow discharges for $d_l = 14$ cm. They are composed of PP of size $\phi \sim 32$ nm, of the same size range than in **Fig. 1**. They are still visible and linked either by van der Waals forces or by covalent carbon bonding. The agglomeration observed here in addition to the case of **Fig. 1** (in both cases the time discharge is 10 min) can be due to a difference in the glow plasma electrical field, induced by a higher electrode gap (14 cm). This could have influence in the PP drift velocity, favoring their agglomeration by coulomb attraction, if one assume that as long as their size remains small, their surface electric charge fluctuates, being alternatively positive and negative¹⁰.

A systematic study of the growth size scaling laws as a function of the discharge duration for $d_{0,l} = 5 - 14$ cm is under way and will be correlated to the plasma characteristics.

The SEM picture of **Fig.3** gives a carbonaceous agglomerate example coming from weakly adherent deposits on the upper surface of the TPL of TS⁷. The SEM analysis, performed in the range of 0.5-200 μm , shows the presence of micrometric flakes of a porous nature. In some of these fragments, the highest magnifications show agglomerates of various size and shape composed of small particulates of various size (**Fig. 3**).

b) Structural analysis between laboratory and TS dust samples

The surface structure and chemical bonds of dust samples coming from the glow discharges (*Lab sample*) and from the LPT surface of TS (*LPT sample*) have been investigated using XANES analysis^{11,7}. The experiments were performed in LURE (Super ACO storage ring, Orsay, France) on the SA72 beam line equipped with a high-energy toroidal grating monochromator providing a resolution of ~ 300 meV at the C K edge, in the total electron yield detection mode. In both cases, no significant angular signal dependence

was observed indicating that the samples are not oriented. A comparison of the two XANES spectra is displayed in **Fig. 4-a**).

In the case of the *TPL sample*, one observes a peak at 285 eV, signature of the C 1s $\rightarrow \pi^*$ transition of the graphite hybridization (sp^2). The continuum structures above 290 eV result from excitonic excitation (291.7 eV) and multiple scattering (MS) resonances within the carbon atoms, in the graphite structure¹¹. The small peak at 288.5 eV is assigned to (C-H)^{*} bonds, likely to be due to the cyclohexane solvent used to stick the powder on the sample holder. This spectrum is therefore characteristic of disordered graphite-crystallites of a few nm length¹².

In the case of the *Lab sample*, the characteristic resonance of unconjugated $sp^2 \pi^*$ (C=C) bond is observed at 284.7 eV. The peaks at 286 eV and 288.5 eV give respectively, the energy position of the π^* (C=O) and σ^* (C-H) transitions. The presence of these elements could be due to air exposure, indicating a high surface reactivity. The continuum region above 290 eV is made of two broad transitions centered around 292 eV and 300 eV, assigned to the σ^* resonances in sp^3 (C-C) and sp^2 (C=C) hybridization states, respectively. These characteristics show that the grains produced in glow discharges contains some unsaturated C=C groups resulting either from C=C defects in the carbon lattice or from the presence of some dispersed aromatic cycles and also contains tetra-coordinated carbon.

The differences observed between the spectra of **Fig. 4-a**) can be explained by a difference of the plasma conditions from which they originate. In particular, the neutral and ion temperatures in which the precursor growth processes take place are different. In the glow discharges for instance, the ion and neutral argon temperatures are smaller than 100°C and the particulates are collected on the anode surface never exceeding 80°C. In TS, for long shocks, because of a weekly adherence of the dust on the TPL surface, their temperature can be higher⁸ than 450°C, heating effect producing carbonization (aromatic cycle formation).

In order to induce some carbonization, both samples were heated ex-situ, to about 450°C. As displayed in **Fig. 4-b**), the σ^* (C-H) signal disappeared in both cases as well as the resonance π^* (C=O) in the *Lab sample* spectrum. In this latter case, the heating also induced a structural change. The shift of the π^* (C=C) from 284.7 to 285 eV clearly shows that the formation of aromatic cycles occurred. The intensity of the π^* (C=C) peak also increased when compared to the continuum at higher energies, showing that numerous carbons, initially in a sp^3 configuration, underwent cyclization. The heating also changed the structure of the continuum region which presents now an overall shape close to the *TPL sample*. However, the

absence of MS oscillations and of the excitonic structure shows that the dust sample carbonization was limited to a spatial range probably smaller than one nm¹².

4- Conclusion

In order to understand the dust formation mechanisms from the condensation of carbon vapors, different situations are analyzed. The conditions of glow discharges with carbon vapor injected by graphite electrode sputtering give rise to spherical primary particles of size in the nanometer scale range. The low temperature of the ions and the neutral, could explain their amorphous structure where the tetra coordinated carbon is dominant. The dust sample taken on the TPL surface of Tore Supra where incident heat loads are high shows a disordered graphitic signature likely to be correlated to the high temperature they can reached because of their weak adherence on the TPL element.

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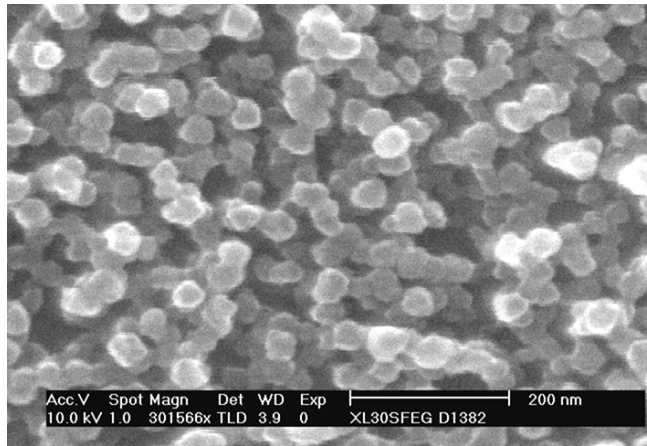


Fig. 1: Primary particles produced after several successive 10 min glow discharges. The distance between the electrode is $d_0 = 5$ cm. The primary particle size is $\phi \sim (38 \pm 5)$ nm.

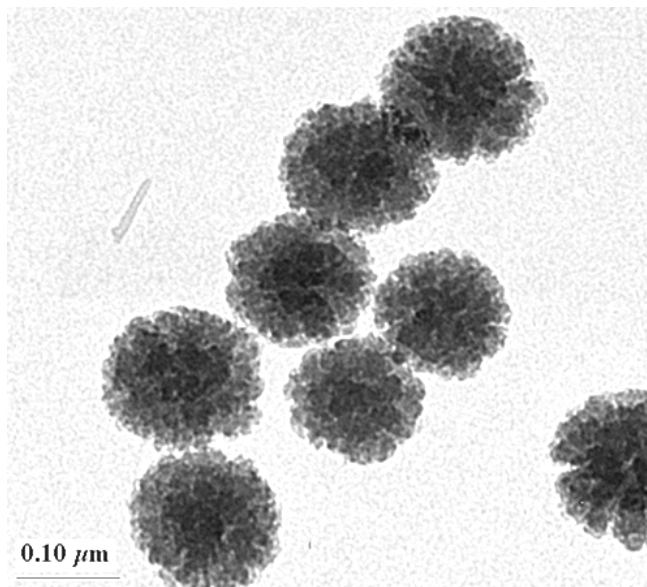


Fig. 2: Spherical aggregates of size $\Phi \sim 190$ nm, produced after several successive 10 min glow discharges. The distance between the electrode is $d_1 = 14$ cm. Primary particles of size $\phi \sim 32$ nm are visible.

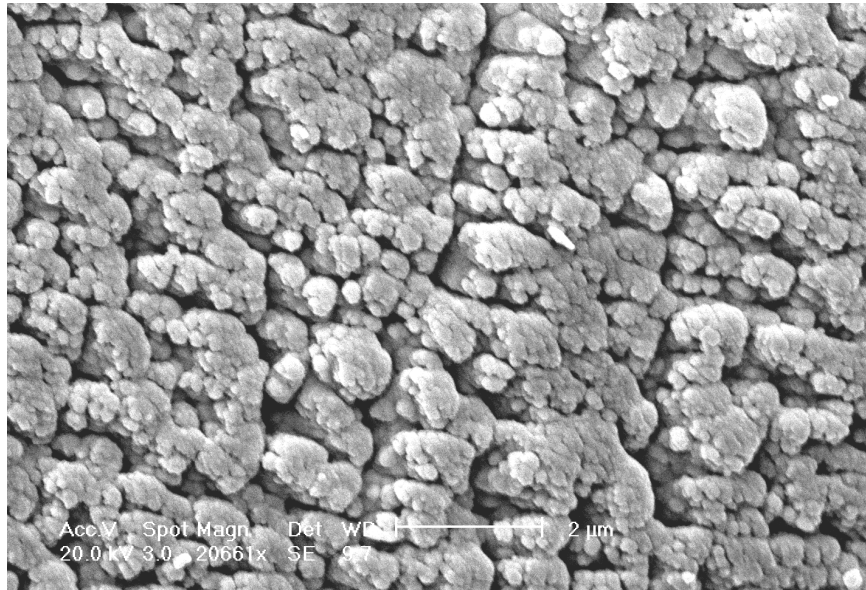


Fig. 3: Agglomerates of different shape and size coming from weakly adherent deposits on the upper surface of the toroidal pumped limiter of Tore Supra.

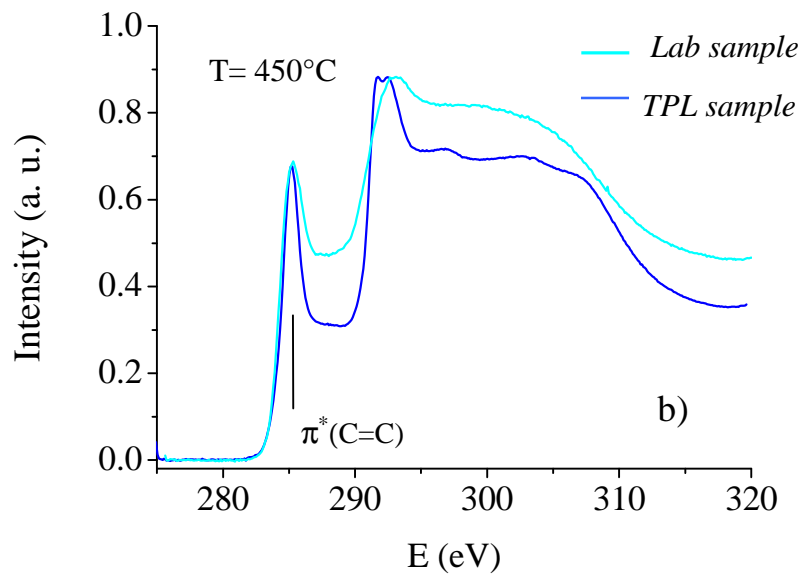
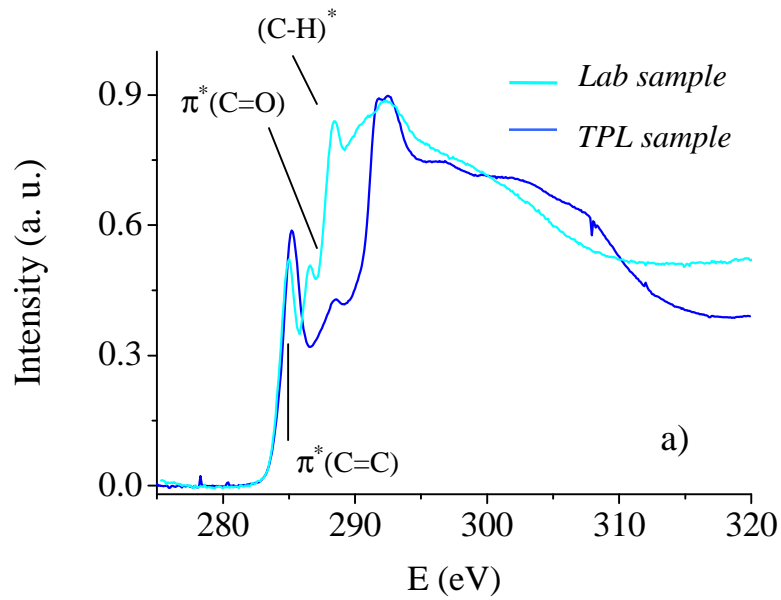


Fig. 4 a), b): XANES C K-edge spectra of a dust sample produced in a glow discharge - *Lab sample*- and collected on the surface of the toroidal pumped limiter of Tore Supra - *TPL sample*- **a)** without heat. **b)** ex-situ heating at 450°.