Study of Dust Morphology, Composition and Surface Growth under ITER-relevant Energy Load in Plasma Gun QSPA-facility

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Abstract.

The experimental date on structure, chemical and X-ray analysis of re-deposited material from eroded CFC and W macrobrush plates designed and manufactured in EU under ITER ELMs and disruptions heat loads are presented. Experiments were realized in plasma gun QSPA facility. Each targets were exposed to a large number of repetitive pulse of heat loads 0.5-2.0 MJ/m^2 with 0.5 msec time duration.

The principal result of impact I type ELMs-like plasma on material was in formation the films, consisted of the dust particles with spherical shapes, typical dimensions of 0.02-2.0 mkm and fractal surface structures (“cauliflower”). In dust particles the Auger spectrometer/SIMS analysis found out many of elements from vacuum chamber surface. X-ray crystal analysis show that tungsten re-eroded in the form of tungsten-carbide (nearby 70%). The analysis of small angle scattering gives “halo” picture, which indicate of 10-20 nm clusters. The calculation for ITER of sorption surfaces (SSA) of dust particles are presented.

1. Introduction

The existence of dust particles inside of a plasma chamber and divertor of ITER has been identified as a serious issue for the development of fusion energy [1]. From experience in existing fusion machines or experiments simulating plasma-wall interactions it is assumed that during ITER lifetime, dust and flakes will be produced due to the ELMs and disruptions interaction with the plasma facing components - Be, W, CFC [2]. The eroded materials during evaporation, melt layer losses with splashed metal droplets and formation of surface cracking with destroying of detached grains tend to re-deposition in dust and layers of stratified mixed materials. Based on the date on carbon dusts from various tokamaks, it has been accepted that the dusts accumulated during ITER operation will have a mass-median diameter of 0.5-2 μm [3], while there is clear evidence that dust produced in some fusion facilities have a nanoscale dimensions [4], [5]. The growth of particles by condensation of the vaporized material lead to fractal structure for the most part. Such particles can be porous with extremely high specific sorption surface (SSA) [6], can have complex material composition Be-W-C and will be activated, tritiated and chemically reactive and toxic. Tungsten is the most hazardous of the materials expected to contribute to the dust dose in addition to expected tritium sorption by fractal dust. The necessity of study fractal structure and growth phenomenon determined not only by the dependence of mechanical, magnetic and electrical properties from the nano and microstructure of material but the absence of estimation plasma interaction with porous deposits in existent simulation.

2. Relationship of experiment to ITER

The investigation of dust particles origination from erosion of CFC and tungsten macrobrush plates was undertaken in frames of joint EU-RF collaborative experiment. Castellated divertor plates were manufactured in Plansee AG (Austria) according to the ITER divertor target specifications [7]. These plates have been exposed to ITER type I ELM loads at the plasma gun facility QSPA at the TRINITI [8]. The energy of the ELM-like loads consists of 1-1.5 MJ/m^2, the duration was 0.5 ms. During the experiments, the macrobrushes were preheated to the temperature of 500 ^{0}C. The surface inclination was 30^0 to the plasma motion. For such energy loads the evaporation is estimated as the major mechanism for W macrobrush erosion and sublimation - for CFC macrobrush [9]. The estimated collector temperature was ≈ 300 ^{0}C. The location of dust collectors meet the requirements of analysis evaporated material spread both in direction of plasma movement and in perpendicular direction. The analysis of erosion products was made after series of, typically, ~ 100 shots. Scanning electron, tunnelling (STM) and atomic-force (AFM) microscopes were used as basic diagnostics for dust analysis.

3. Nanodust morphology. Agglomeration

Intensive surface erosion by plasma in tokamaks and many other fusion facilities lead to formation the great deal of amorphous flakes with irregular structure, which can destroyed easily into dust. As example T-10 hydrocarbon flakes has smooth surface with globular

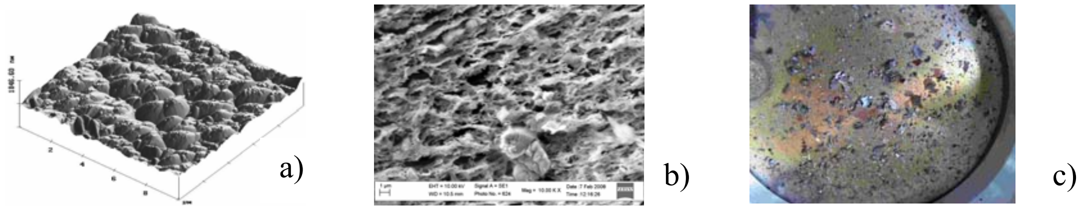


Fig.1. Tokamak T-10. a) hydrocarbon globular film, b) hydrocarbon porous film, c) dust/debris on the vacuum valve

structure (Fig 1a) or high porous body material structure (Fig 1b). The debris which cover the T-10 tokamak flange surface (Fig.1c) has typical dimension from 0.1 μm to some mm.

A radical difference of impact type I ELM-like plasma on W and CFC macrobrush was in formation the films, consisted of a large number of dust particles with spherical shapes (Fig.2). Spherical particles have "cauliflower" structures both for carbon and tungsten. In other words they were formed by agglomeration of small clusters, which in turn consisted of clusters with even smaller size, etc. The maximum size of the spherical dust particles is 2-3 μm, but tungsten dust in contrast to carbon dust keeps on agglomeration into elongated clusters ~ 5-15 μm lengthwise, which exhibit a tendency to roll up into toroids (fig 5a).

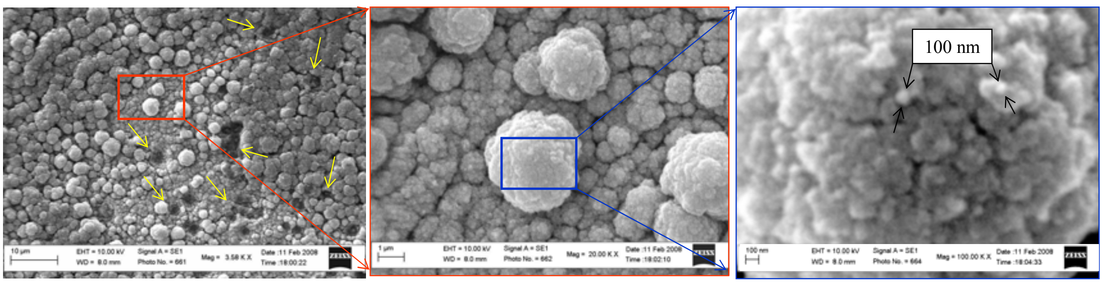


Fig.2. QSPA. Dust particles after W target erosion with 1.5 MJ/m^2 energy load.

Further investigations by STM/AFM found out that the “cauliflower” structure in reality consist of the nanoparticles with dimensions of 20-50 nm and less. The structure consist of the nanoparticles have to big sorption surface.

The investigation of collectors with dust show that they coated non-uniformly by material. The area dotted with big 2-10 μm clusters adjoined with parts covered by only small (0.1-0.5) dust particles. The analysis of material deposit with increasing of plasma shots denote two parallel processes – agglomeration of nanoparticles into “cauliflower” dust and dust tighten/motion/diffusion into circular flat “islands / pancakes” or “tongues of flame” structure (Fig.3), scattered between separate dust clusters. After big number of shots (>100) on the collector surface formed the dense, coral-like coating from the dust particles. Since spherical clusters can migrate the bond with the surface can not be durable. Thereto may denote the hole from dust on Fig. 2a (yellow indicator).

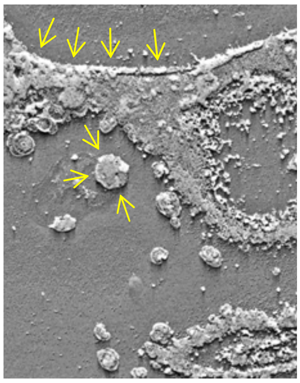


Fig.3 Dust tighten on collector surface

4. Dust composition

Analysis of chemical composition of coral-like tungsten film consist of dust particles was carry out in FESEM by Auger spectrometer with spatial resolution near 1μm. Many elements of material, which used in previous experiments, the elements of SS vacuum chamber and materials of diagnostics were found out on the film surface (fig. 4a). But for all that in deeper layers there is much less impurities (fig 4b). To all appearance there is evaporation of thin layer of wall material and films, covered the vacuum chamber surface, with further mixing this material with tungsten during plasma load on macrobrush plate.

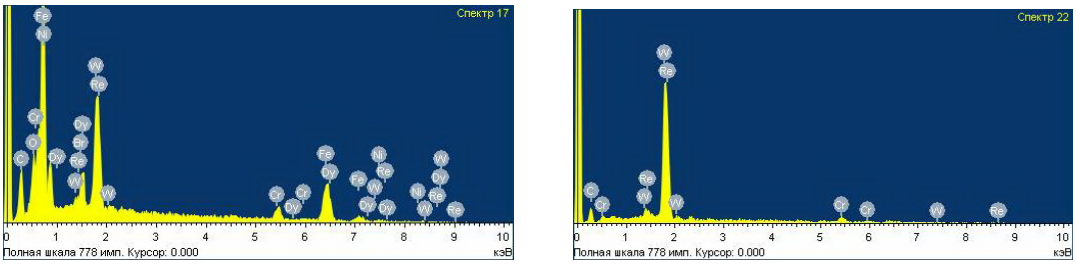


Fig.4. Auger spectrum of W dust. a) on the surface, b) in deeper layer

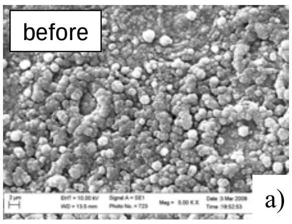
X-ray analysis of film deposited during tungsten erosion by 1.5 MJ/m^2 pulse plasma load was carried out by diffractometer used CuKα-monochromatic radiation. The crystallogram halo of small angle radiation scattering gives the value of clusters dimension near 10-20 nm.

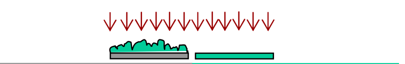
Results of phase analysis presents in Table 1. Basically deposited tungsten settle in form of tungsten carbide. The pure W keep only ~ 30%. Obviously during pulse energy load enough carbon evaporated for formation the tungsten carbides.

W	WC1-x	W_2C	WC	FeW_3C	Fe_6W_6C	Graphite
2	1	1	1	1	1	2

5. Surface modification

It would be mentioned that if structure consist of the 10-20 nm particles the sorption surface area (SSA), and accordingly surface energy, usually have to be very high. Especially it relate to fractal films. The experiments on simultaneous sputtering of W fractal dusty film and polycrystalline W film by Ar^+ beam (3 keV, 3 mA/cm^2) was made to indicate the properties of the big surface energy of fractal film. Experiment find out the weight loss (or





W (SS collector)	Polycrystalline W
ΔP = 1.8 mg $Y \approx$ 0.20 at/ion	ΔP = 9.5 mg $Y \approx$ 1.20 at/ion

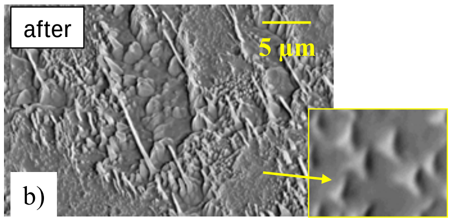
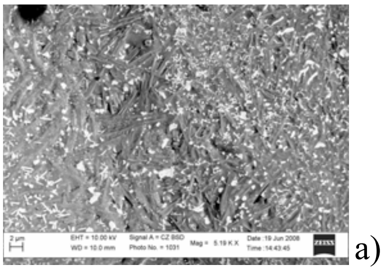


Fig.5. Ar^+ sputtering of W dusty film. a) before modification (b) after modification

sputtering yield) of such film 6 times less then that of polycrystalline W film. For all the surface structure of polycrystalline W film didn't changed much, the surface structure of W fractal film changed dramatically. There are appeared big uniform areas with ample evidence of "liquid melt" and circular, smooth particles equal diameters and high density surface whiskers. Also the third order symmetry axis typical for monocrystalline tungsten appeared on the surface of "melt" (Fig.5b). At first sight It is unusual that evident transition of material from fractal phase to crystalline phase take place with specific Ar^+ beam power 5 W/cm^2 only, although well known W fusion heat equals ≈ 200 J/g. But if suppose, that surface energy of fractal clusters convert into fusion heat, than such transition can supply with junction of clusters of some nanometer dimensions, which is in good agreement with experimental dates.



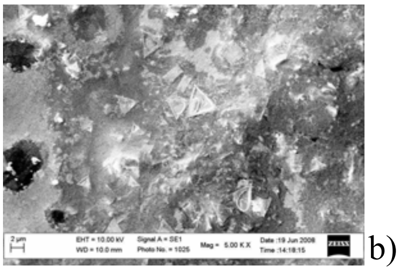


Fig.6. Surface morphology of collectors, which placed on the direction of plasma/material jet.

Similar structures were finding on collectors, which placed inside QSPA vacuum chamber, along the lines of plasma and erosion products motion. These collectors were installing over the macrobrus plates. Fig. 6a show columnar structure, covered by amorphous paste. On Fig.6b the collector surface spangled by crystall. Chemical analysis of crystal finds only tungsten and carbon. Obviously the energy in plasma stream enough for modification of material which carry by plasma jet.

6. Dust particles distribution. Fractality

Generally the surface of collector take out from the QSPA vacuum chamber are intersperse by dust particles having fractal ("cauliflower") structures. As it was shown earlier [10] the sputtering of W and CFC by ITER ELM I type plasma energy load with 1.0 ÷ 1.5 MJ/m^2 result in power-law dependence of particle size distribution i.e. $N \sim r^{-\alpha}$ with $\alpha = 2.2 – 2.3$ in the characteristic particles size range 0.1 ÷ 2 μm. Additional investigations of particle size distribution by STM/AFM in the range of 20-100 nm found out the power-law dependence can be extend up to

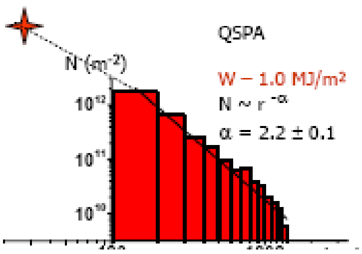


Fig.7. QSPA-facility dust size distribution

20 nm range (Fig.7). Further experiments verify such distribution. Similar power-law dependence of particle size distribution with the same power $\alpha = 2.3$ in the range of 1 nm -10 µm found out in stellarator LHD [4].

The same results were obtained in tokamak T-10 with high power load to the graphite limiter. In this regime half of the tokamak heating power came into the small inner part of circular limiter providing the specific power load ~ 50 MJ/m^2 during 0.5 sec. [11]. This lead to increasing surface temperature up to 2000 ^{0}C, strong sublimation of graphite under intensive arcing, dust appearances with initial dimension near 25 nm and further agglomeration. And the same power-law dependence of particle size distribution i.e. $N \sim r^{-\alpha}$ with $\alpha = 2.3$ was obtained in the range 50 – 200 nm. The results from some facilities [4][12] reveals the similar dependence. It is significant that the power-law dependence with the fractional power describe the fractal structures.

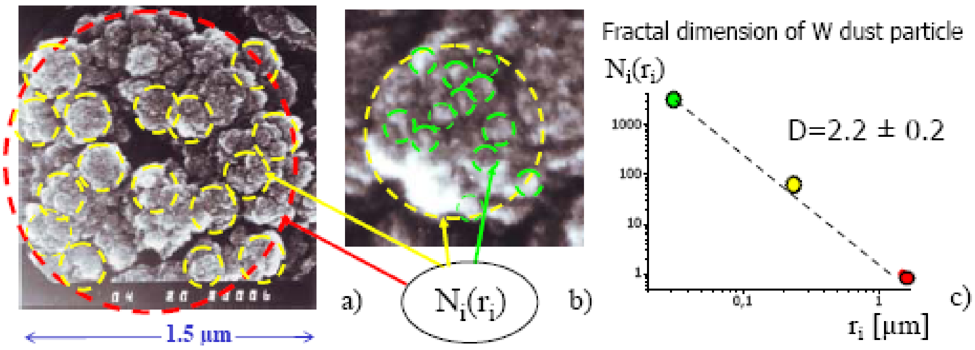


Fig.8. QSPA dust hierarchical structure (a,b) and fractal dimension (c).

The analysis of individual QSPA dust particles or agglomerates found out their "cauliflower" or fractal structure. The fractal dimension can be viewed as a relative measure of a complexity, or as an index of the scale-dependency of a pattern. To estimate the fractal dimension of the object, we use the box-counting method (see, e.g., [13]). The fractal dimension of a fractal object contained in Euclidean space with Euclidean dimension of E is estimated as following: for any $r > 0$, let N(r) be the minimum number of E-dimensional cubes of side-length r needed to cover a fractal object. If there is a number D so that $N(r) \sim 1/r^D$ as $r \to 0$, the fractal dimension of the fractal object is D. An estimate of the fractal dimension D is obtained by calculating the slope of plot log(N(r)) vs. log(1/r). For Euclidean object, D is an integer (Euclidean dimension). For a fractal object, D is not an integer. We use this procedure to estimate a fractal dimension of a film volume. On the micrograph in Fig.8 several hierarchical levels of granules (from 20 nm up to 2 µm) are observed. We found N(r), which is the density of granules of radius r on the micrograph. In the log–log plot, the experimental data could be fitted by a linear function . From this plot, fractal dimension D was estimated as 2.2. Error bars are estimated from the fitting of the data by a linear function using χ^2 method. For many films from the QSPA facility, the fractal dimension D is in the range between 2.15 and 2.3. It is essential to pay attention to the fact of proximity the values of α - which describe distribution of total number of dust particles and fractal dimension – D of single particles. Thus, if fractal cluster with the size r_0 consist of the particles with size a_0, then the numbers of particles inside the r_0: $\mathbf{N_{r0} = (r_0 / a_0)^D}$.

7. Mechanism of fractal dust growth

In general, the formation of a fractal landscape in the nature is often a process of erosion or a deposition. Scaling concepts have become one of the main tools in the study of growth processes in condensed matter. A large number of numerical models of a surface growth, of varying levels of sophistication, has been developed [14]. DLA (*Diffusion-limited aggregation*) model [15] [16] is a very simple model that is widely applicable for a description of a lot of natural phenomena.

In the DLA model, the process starts from a fixed seed particle. Introducing another particle at a large distance from the seed, it walks randomly until sticking to the seed or escaping to infinity. This aggregation process is repeated many times generating a cluster with a fractal structure. To demonstrate the importance of diffusion statistics for the morphology of the surface growth, we have performed numerical simulation of the surface growth in DLA. A result of the DLA fractal growth in 2D plane is shown in Fig. 9. We started from several seeds. The trajectories of attached particles were simulated by numerical

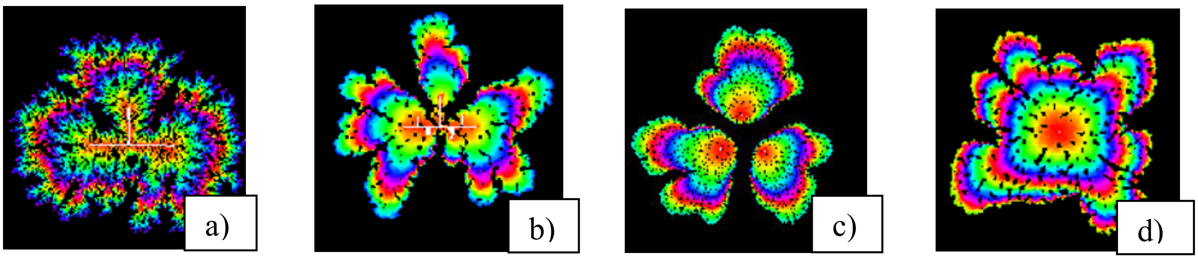


Fig.9. DLA with a variation in the statistics of the deposited particle diffusion. Simulation with the Hurst exponent of diffusion: (a) H=0.5, (b,c,d) H= 0.8.

approximation of the stochastic integral [17]. Two cases of different statistics of diffusion have been used: Brownian motion (classic diffusion) with the Hurst exponent of H=0.5 (Fig. 9a), FBM (superdiffusion) with the Hurst exponent H=0.8 (Fig. 9b,c,d). This result illustrates how the cluster texture depends on the diffusion statistics of attached particles. Normal diffusion with H=0.5 generates a tree-like structure. A cauliflower shape is generated only in the aggregation of particles attached through FBM with H>0.5. The parameters of DLA model – sticking probability and motion onto the cluster brunch – mach influence on fractal cluster shape. The image (Fig. 9c) is qualitatively compatible with the texture of QSPA dust.

Feature of topography of dusty films growth during divertor plate erosion by high pulse energy load also can explain on the basis of structures growth from the single atoms deposited on a surface within the limits of the theory of crystals growth, and in particular, theory of crystal seed formation [18]. Deposited atoms on a surface make two-dimensional gas. Concentration such addatoms on a surface C with in the assumption of absence of their removal from a surface (evaporation or sputtering) is defined by a flux of atoms onto the surface from vacuum - q, leaving (diffusion) on flowing and association into the clusters.

$$\partial C/\partial t = q - DC/aL - DC\cdot\Sigma n_i ,$$

where D – surface diffusion constant of addatoms, L - distance to a drain, a – the atom size, n_i – concentration of clusters consist from i atoms. Addatom association in clusters occurs, either due to fluctuating associations of several atoms till the size of a critical seed, or at the joining to already available clusters or to impurity which serve as the centres of seed origin. The basic assumption which accept for an explanation of the developed topography of a surface is **accumulation of impurity** on a growing dust/film surface. The impurity can play a role of the condensation centre round which the crystal seed is formed. Impurity atoms accumulation on a surface has no saturation. Impurity atoms "emerge" on a surface for the account of diffusion flux towards a surface caused by a temperature gradient and high temperature. Impurity with big difference of dimension from own dust atoms size generate cluster with a screw dislocation which grows as a whisker. The same impurity atoms are account for whisker bifurcation according to fractal law and formations of the "cauliflower" type structure. The elements of “cauliflower” structure are often similar not to a stick but on rice grain or even on a ball. It is possible to explain the stick diameter growth by deposited atom sedimentation. The developed structure is caused by addatoms migration on a surface to the **hottest places of structure.** This creates fractal structures growth by the mechanism similar to the mechanism formation of "viscous fingers» or the DLA.

8. Extrapolation to ITER

One of the main dangers in ITER is films/dust/deposits growth with highly developed surface that may lead to high absorption of deuterium and tritium. The proposed mechanism of surface particles agglomeration into the fractal clusters can create serious problems for ITER. To suppose that the mechanism of divertor plates material redeposition in ITER has the same nature as the QSPA macrobrush material redeposition, the Specific Surface Area (SSA) can be estimated. For the solid-state particles no difference between SSA estimated

$$\text{SSA} = \; S_0/\rho V = \int_{a_0}^{r_{\max}} Ar^{-\alpha} 4\pi a_0^{\,2} (\frac{r}{a_0})^D dr \Big/ \rho \int_{a_0}^{r_{\max}} Ar^{-\alpha} \frac{4}{3}\pi a_0^{\,3} (\frac{r}{a_0})^D dr = \frac{3}{\rho a_0}$$

from existent ITER log-normal dust distribution and power-law dust distribution, where SSA depends on highest possible cluster dimension $\boldsymbol{r_0}$. But quite the contrary, if dust clusters have **power-law** distribution and **fractal** structure, then SSA depends on minimum dust size $\boldsymbol{a_0}$. Assuming in accordance with the experiments $\boldsymbol{a_0} \approx 10$ nm it results in SSA ≈ 250 m^2/g for carbon and 30 m^2/g for tungsten that is **≈ 60 times higher** of existent calculations of ITER SSA (≈ 4 m^2/g for carbon). Thus, in ITER it is necessary to expect tritium accumulation in redeposited films much times above, than is accepted in the modern project.

Fractal structures have the big specific area of a dust surface and consequently the big reserve of specific energy comparable to corresponding specific energy of explosives. At film structure compression specific surface energy of system decreases. At such process the overflow energy utilize on system heating and structure destruction. Explosion of these structures is possible (for example, at local thermal loading) with formation of a considerable quantity of dust particles not only micro, but also nanoscale sizes. Obviously the nanoparticles mass fraction in comparison with the microparticles can be high enough as decomposition of a small particle demands smaller energy.

Fractal films can possess the increased electron emission which influences on energy and particles balance in divertor. Such mechanism – the amplification of secondary electron emission in 100 times and reduction of work function is observed in a number of experiments.

It is necessary to give a great attention to a problem of the ITER first wall clearing from the metal porous films containing tritium. The surface modification of porous films can "immure" tritium in deep layers.

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