Scale Symmetry of Stochastic Surface Clustering under Plasma Influence in Fusion Devices

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Abstract: Titanium, tungsten, carbon, lithium, and beryllium surface structure were analyzed after plasma irradiation in fusion devices. Exceptional extreme high-temperature plasma load in fusion devices leads to specific surface clustering. It is strictly different from any other conditions of material’s clustering. The hierarchical granularity with cauliflower-like shape and surface self-similarity have been observed. Height’s distribution is deviated from the Gaussian function. The relief roughness differs qualitatively from the ordinary Brownian surface and from clustering under other conditions. In fusion devices, the specific conditions regulate material surface clustering faced to plasma. Ions and clusters melt on the surface and move under the effect of stochastic electromagnetic field driven by the near-wall turbulent plasma. In such a process, long-term correlations lead to the growth of surface with a self-similar structure. The multiscale synergistic effects influence the self-similarity–fractal growth from nanometers to millimeters. Experimental results illustrate universality of stochastic clustering of materials irradiated with plasma in fusion devices.

Keywords: scale symmetry; stochastic clustering; plasma–surface interaction; fractal growth

1. Introduction

In fusion devices, the effect of high-temperature plasma on the material surface causes intense erosion of plasma-facing materials, redeposition of eroded matter and a significant change in the surface structure. As a result, surface layers with a specific stochastic topography and a hierarchy of material granularity are growing at scales ranging from nanometers to millimeters [1,2]. The role of nanoscales is important in dendritic growth providing various pathways for anisotropic overgrowth and aggregation-based growth leading to branched structures or hierarchical granularity of fractal topology. In such growth, the reduction in surface energy is achieved by eliminating attached areas of surfaces, which provides a strong thermodynamic driving force for particle (ions and clusters) interaction and attachment. If nanoparticles and clusters are diffused by stochastic motion, they are likely to aggregate in a diffusion-controlled process. Diffusion-limited aggregation (DLA) process regulates the growth of large-scale clusters leading to scale invariance topology of structure formation over broadband scales from nanoscales to macroscales. The properties of the scale invariance relate with the formation of a percolation cluster of defects and dissipative structures. Scale invariance (self-similarity) in the structure and self-organization of dissipative structures at the nano- and meso-scales can determine the universal features of solid material at the macro-scale.

Due to a strong plasma-surface interaction (PSI) in fusion devices, the dynamics of agglomerated particles and clusters on the plasma-facing material is controlled by several driven and damping growth processes. These elementary processes influence simultaneously producing synergistic effects described by the theoretical models [3] of surface growth instability. Near-wall plasma turbulence in fusion devices produces the fluctuating electric field, which leads to stochastic motion of deposited ions and clusters.
resulting in the fractal growth of the surface. Intermittent turbulent fluctuations of electric field with non-Gaussian statistics [4] produce specific dynamics of particles including Levy flights and long-range correlations. The statistics of surface relief heights is inherited from non-Gaussian statistics of such plasma turbulence producing a surface particular shape with unique scale symmetry (e.g., cauliflower-like). Scale symmetry describing the structure of materials under extreme plasma exposure are needed to develop the fundamental concepts of the physical properties of matter. It is important to study the micro-, meso-, and macro-scales in solid materials, the properties of the percolation structure that determine the critical concentration of fracture centers under extreme conditions.

Therefore, in order to identify the universality of a formation and the behavior of solids with a fractal structure of drastically different behavior from systems close to equilibrium, it is important to study in detail the scale invariance features, starting from nanoscales. Such information is necessary to the study of solid materials with fractal structure under extreme loads and to develop new fractal materials technology attractive for nuclear and chemical applications.

In this paper, the characterization of materials irradiated with plasma in fusion devices regarding a scale symmetry of the structure is described.

2. Materials Irradiated with Plasma in Fusion Devices

In several fusion devices, materials (titanium, tungsten, lithium, carbon, and beryllium) of initial smooth surface and crystalline structure were irradiated with plasma. A stochastic surface with different scales of hierarchical granularity is formed on the tested samples. This surface has a rough stochastic relief over broadband scales from nanometers to millimeters. In particular, the cauliflower-like hierarchic shape is formed.

Material specimens were irradiated in fusion plasma devices: the PLM device [5], the T-10 tokamak [6], and the quasi-stationary plasma accelerators—QSPA-T [7] and QSPA-Be [8]. The PLM plasma device [5] is a multicusp linear trap with a steady-state plasma discharge providing the powerful steady-state thermal plasma load on tested material. The T-10 tokamak [5] is a medium-scale tokamak with graphite limiters loaded by high-temperature plasma. The QSPA-T and the QSPA-Be facilities are the plasma accelerators to test fusion materials (tungsten and beryllium) with powerful plasma jets. Helium plasma in the PLM device and deuterium plasma in the T-10 tokamak and QSPA were used in experiments. During the experiment, sample materials eroded from the surface of irradiated samples are evaporated into the plasma volume and redeposited on the sample and on the plasma-facing components of the plasma device.

Titanium (Ti) samples of initial smooth surface were irradiated with helium plasma in PLM for 200 min [9]. The heat load on titanium samples was of 0.5–1 MW/m². The plasma irradiation leads to a formation of hierarchical granularity of the plasma-facing surface, Figure 1.

Tungsten samples from the T-10 tokamak, Figure 2, were obtained in experiments with tungsten limiter, see the description of the experiment and samples in [10].

Carbon samples, Figure 3, have been collected on the vessel of the T-10 tokamak after series of experiments with graphite limiter (see [2,11,12]). The carbon layers were grown as deposition of limiter materials eroded under plasma load of 1–10 MW/m² during 1000 discharges in the T-10 tokamak of more than 1000 s total duration.

Lithium samples were formed after plasma irradiation in the T-10 tokamak and PLM plasma device [13,14]. The lithium samples as layers of 0.5–2 mm thickness are formed on in-vessel components of the T-10 tokamak after experimental campaign described in [13,15]. In these experiments, the liquid lithium of capillary porous system evaporated under a powerful plasma load in plasma discharges. The evaporated materials redeposited on the surface. The reactivity of lithium with oxygen and carbon in plasma resulted in deposited materials with hierarchical granularity, see [13,14]. These lithium samples were subsequently irradiated with plasma in the PLM plasma device, see [13,14]. Under the steady-state plasma load of 200 min in PLM, surface structures with hierarchical granularity
were grown on scales of 0.5–500 microns [13], Figure 4. The cauliflower-like surface relief is similar to the morphology of hydrocarbon deposits observed recently in plasma devices tokamaks [11,12,16].

Figure 1. SEM micrograph of titanium sample surface irradiated with plasma in PLM plasma device.

Figure 2. SEM micrograph of tungsten sample surface irradiated with plasma in the T–10 tokamak.

Figure 3. SEM micrograph of carbon surface of the sample from the T–10 tokamak.
Figure 4. SEM micrograph of lithium surface of the sample from the T–10 tokamak and subsequently irradiated with plasma in the PLM plasma device.

A reference industrial powder of lithium carbonate Li$_2$CO$_3$ was tested in PLM with the same plasma parameters to compare with irradiated samples from the T-10. The plasma irradiation of the reference powder resulted in the self-similar surface structure, Figure 5.

The tungsten plates were irradiated with powerful plasma in the QSPA-T facility [1,16,17]. The energy of the plasma load was up to 1 MJ/m$^2$ during one pulse of 0.5 ms duration, Figure 6, and with 1 MJ/m$^2$ plasma load during 100 pulses of 0.5 ms, Figure 7 (simulation of edge localized modes—ELMs—in a large tokamak). The stochastic surface with granularity is grown on the tungsten sample, Figures 6 and 7.

The beryllium sample were irradiated with ten plasma jets of 0.5 ms duration in the QSPA-Be [8], Figure 8. The plasma pulse energy of up to 2 MJ/m$^2$ was used to process the material surface.

Two reference samples were produced under conditions distinguished from those described above. The stochastic roughness of these reference samples was formed due primarily to only one mechanism. Reference sample of molybdenum produced in a magnetron low-temperature plasma (the sample is described in [1,18]) was analyzed to compare its structure with above samples from fusion devices. Ballistic deposition of the
erosion products from plasma on the surface is a dominant mechanism of the surface growth in the magnetron discharge.

Additionally, the industrial steel casting with the untreated rough surface formed after solidification in a casting mold was used for comparative analysis. This sample (see details in [1]) is a typical example of solidified melt of metal with ordinary surface roughness specified in industry.

Figure 6. SEM micrograph of tungsten sample surface irradiated with plasma in the QSPA–T plasma facility.

Figure 7. SEM micrograph of tungsten sample surface irradiated with plasma in the QSPA–T plasma facility.

Figure 8. SEM micrograph of beryllium sample surface irradiated with plasma in the QSPA–Be plasma facility.
3. Methods of Surface Analysis

The scanning electron microscope (SEM) was used to produce the image (micrograph) of a sample by scanning the surface. For the titanium, lithium, beryllium, and carbon samples (Figures 1–8) the SEM image resolution was up to 20 nm.

For the titanium sample, the measured relief heights (Figure 9) demonstrated intermittent behavior with non-trivial stochasticity. For the tungsten sample (Figure 10), profiles of 0.2 mm length were measured with the base resolution of 20 nm (see the description in [2]).

Figure 9. Heights profile of a titanium surface after plasma irradiation in the PLM device.

Figure 10. Heights profile of a carbon surface after plasma irradiation in the T–10 tokamak.

The probability distribution function (PDF) of the relief is formed as the normalized histogram of the heights (see [2,19]). The PDF of experimental profiles was compared with the Gaussian law to characterize specific property of a roughness. The Gaussian PDF is typically observed for objects with the trivial roughness like the Brownian surface (see examples in [15]).

To characterize inhomogeneous statistical feature of the relief, the Hurst exponent, scaling of the structure function and multifractal spectra (see, e.g., [2,15,20]) are usually used. The Hurst exponent characterizes self-similarity under the assumption that its properties are independent on the scale of observation. The Hurst exponent allows comparing the structure with the known theoretical models of stochastic objects.

Statistical self-similarity is considered when changes on scales $l$ and $\lambda l$ are described by the cascade rule, $\delta_{\lambda l}y(x) = W_\lambda \delta_l y(x)$, where $W_\lambda$ is a random variable depending only
on $\lambda$, $\delta y = y(x + l) - y(x)$. For self-similar function $y(x)$, the probability distribution function $P_l(\delta y)$ for increments is $P_l(\delta y) = \lambda^H P_\lambda(\lambda^H \delta y)$ and is characterized by the Hurst exponent $H$. A persistent behavior (trend) is described by the value of $H > 0.5$, like irregular stochastic clustering with hierarchical granularity (fractality), e.g., the cauliflower-like. The procedure of estimating the Hurst exponent $H$ is described in [15].

To describe inhomogeneous stochastic structure, a dependence of the PDFs for $\delta y$ was analyzed. For multifractal object such PDFs change the shape from the Gaussian at large scales $l$ to a non-Gaussian at small scales $l$ (see an example in [2]).

The spectrum of dimensions $D(h)$ was used to characterize multifractal object with a set of the Hölder exponents $h$. The Hölder exponents are the local exponents of scale invariance. $D(h)$ is called the multifractal spectrum or the spectrum of singularities. $D(h)$ for a monofractal object like the Brownian one is a single point with a single Hölder exponent. The procedure of $D(h)$ estimating from experimental data is described in [20].

The method of structure functions $S_q(l)$ (moments of the PDF) (see [20]) was used to analyze the PDF non-homogeneity at various scales. The structure function $S_q(l) = \langle |\delta y|^q \rangle$ was estimated from the statistical averaging $\langle ... \rangle$ with the PDF $P_l(\delta y)$ at the spatial scale $l$. The exponent $\zeta_q$ from a scaling $S_q(l) \sim l^{\zeta_q}$ is useful to analyze a self-similarity [20]. A linear dependence of $\zeta_q$ on $q$ relates with the simplest (trivial) self-similarity, i.e., statistical homogeneity. The object with multifractal statistics (i.e., statistically inhomogeneous) was characterized by the nonlinear dependence of $\zeta_q$ on $q$.

Fractal dimension or Minkowski dimension (known as well as the Minkowski–Bouligand dimension) of the surface structures was estimated analyzing the SEM images by the box-counting method. The number $N(r)$ of boxes of side length $r$ required to cover the SEM image with the set of boxes is a scaling of $N(r) = k r^{-df}$; the fractal dimension was estimated as $d_f = \partial \log(N)/\partial \log(1/r)$.

4. Results of Surface Structure Analysis

The relief heights and SEM images (Figures 1–10) were analyzed to describe scale symmetry properties of stochastic clustering after plasma irradiation in fusion devices. Self-similarity exponents were estimated by methods described above to reveal specific statistical deviation from trivial stochastic relief like the Brownian surface.

For all materials irradiated with plasma in fusion devices, the probability distribution functions of the relief heights are typically not fitted by the Gaussian law, Figures 11 and 12. Figure 13 shows the Gaussian PDF of the reference industrial steel casting surface. This PDF demonstrates the trivial homogeneous statistical properties of the clustering.

![Figure 11. PDF of the profile heights of titanium surface after irradiation with plasma in PLM device. The Gaussian (dotted line) and the Cauchy–Lorentz (solid line) distributions are shown for the comparison.](image-url)
Figure 12. PDF of the profile heights (Figure 10) of carbon surface after irradiation with plasma in Figure 10. tokamak. Surface height increments \( \delta y = y(x + l) - y(x) \), \( l = 19.5 \text{ nm} \) are analyzed. The Gaussian (dotted line) and the Cauchy–Lorentz (red dashed line) distributions are shown for the comparison.

Figure 13. PDF of the profile heights of industrial steel casting surface after solidification. Surface height increments \( \delta y = y(x + l) - y(x) \), \( l = 0.5 \mu \text{m} \) are analyzed. The Gaussian (dotted line) and the Cauchy–Lorentz (solid line) distributions are shown for the comparison.

To describe the statistical scale invariance (a self-similarity) of the surface topology, the Hurst exponent, the structure function scaling and multifractal spectra were estimated. The Hurst exponents for studied reliefs (Table 1) of titanium, tungsten, lithium, carbon, and beryllium were from 0.55 to 0.9 \( (H > 0.5) \) characterizing a persistent trend. It corresponds to stochastic clustering with hierarchical granularity like the cauliflower shape. Hurst exponents from 0.55 to 0.9 correspond to the fractal dimension \( d_f = 2.1-2.45 \) \( (H = 3 - d_f, \text{ see [15]} \).)
Table 1. Fractal dimension (the Minkowski dimension), Hurst exponent $H$ of the surface of samples irradiated with plasma in fusion devices.

| №  | The Sample                                                                 | Fractal Dimension (the Minkowski Dimension) | Hurst Exponent $H$ |
|----|-----------------------------------------------------------------------------|---------------------------------------------|-------------------|
| 1  | Titanium sample surface irradiated with plasma in PLM plasma device         | 2.35                                        | 0.65              |
| 2  | Tungsten sample surface irradiated with plasma in the T-10 tokamak          | 2.2–2.3                                    | 0.8–0.7           |
| 3  | Tungsten sample surface irradiated with plasma in QSPA-T plasma facility    | 2.19–2.3                                   | 0.81              |
| 4  | Lithium sample from the T-10 tokamak and subsequently irradiated with plasma in PLM plasma device | 2.05                                        | 0.95              |
| 5  | Industrial lithium carbonate Li$_2$CO$_3$ irradiated with plasma in PLM plasma device | 2.45                                        | 0.55              |
| 6  | Carbon surface of the sample from the T-10 tokamak.                        | 2.19                                        | 0.80              |
| 7  | Beryllium sample surface irradiated with plasma in QSPA-Be plasma facility | 2.1                                         | 0.9               |

The dependences of the PDFs for the increments $\delta y$ on the scale $l$ were studied to characterize a statistical inhomogeneity of granularity called multifractality. Such PDFs (see an example in [2]) change from the Gaussian at large scales $l$ to a non-Gaussian shape with heavy tails at small scales $l$.

The typical spectra $D(h)$ for the profiles in Figure 10 are shown in Figure 14. The convex bell-like shape spectra $D(h)$ are observed for the samples from fusion devices. Two features of the spectra (broadening and bell-like shape) are typical for multifractal objects in nature (see [20]). The broadening $\Delta h$ of the $D(h)$ are in the range of 0.5–1.2 for studied samples form fusion devices. The spectra $D(h)$ for the relief of reference samples (industrial steel casting and molybdenum) with broadening $\Delta h$ is less than 0.2–0.3 (see [1]) illustrate a deviation of their structure complexity from sample’s structure irradiated with fusion plasma.

![Figure 14](image-url)

For all samples from nuclear fusion devices, structure function calling $\xi_q$ is a nonlinear function of $q$ (Figure 15). The convex shape of $\xi_q$ is similar to the scaling of the log-Poisson
models of a cascade process describing known multifractal objects and processes, e.g., turbulence [4,20].

Figure 15. Scaling $\xi_q$ of structure functions vs. $q$ for the relief of the carbon sample (cf. Figure 10) irradiated with plasma in the T-10 tokamak (crosses). A dependence of $\xi_q$ normalized with $\xi_3$ and subtracted with $q/3$ is shown for illustration of nonlinear behavior, which is typical for objects with multifractal statistically inhomogeneous structure. For the comparison, the linear scaling $q/3$ predicted for simplest (trivial) self-similarity (K41—Kolmogorov’s K41 model [21]) of statistical homogeneity (dashed line), log-Poisson scaling of models SL and BM, see [20], of a cascade process (solid line and point-dashed line) are shown.

The fractal dimension (the Minkowski dimension) of the samples surface were estimated by box-counting method measuring the SEM images (Figures 1–8) complexity. Figure 16 illustrates the box-counting method results for carbon material from the T-10 tokamak. The results of the fractal dimension estimated for the samples studied are in Table 1. The fractal dimension of the surface irradiated with plasma in fusion devices was in the range from 2.05 to 2.35.

Figure 16. Estimation of fractal dimension by the box-counting method. Carbon material irradiated with plasma in the T-10 tokamak (stars). Fitting with power law $N(r) \sim r^{-d_f}$ is shown by a dashed line, estimated fractal dimension $d_f = 2.19$. 
5. Discussion

The interface boundary growth and deposition from the volume to the surface are described in the literature (see, for example, [3]). The reported here experimental data on the structure of material surface support the mechanism of fractal surface growth forming a scale invariance structure. The fractal surface growth was also observed for molecular beam epitaxy, vapor deposition, etc. [3]. Several driven and damping growth mechanisms (elementary processes) on large spatiotemporal scales are involved in the fractal growth. The stochastic clustering of materials irradiated with the high-temperature plasma in fusion devices is mainly caused by collective (synergistic) effect rather than heating itself or exposure to hot plasma reactive species or other single effect (see discussions in [1]).

Estimated fractal dimensions $d_f$ of the plasma irradiated surfaces are in the range of 2.05–2.45, which are comparable with some irregular solids, e.g., metallic glasses ($d_f = 2.31$), silicon aggregates ($d_f = 2.27–2.65$), and quasi-crystals, see [22,23].

In materials irradiated with plasma in fusion devices, a granularity is observed from nanoscales to macroscales (see Figures 9 and 10). It is known that nanoscale structural elements (for example, nanocrystallites) in a nonequilibrium process in a disordered solid, due to their mobility and adaptability, provide scale invariance of the distribution of stress fields at the microscopic and mesoscopic levels. In thin films, under extreme conditions, scale invariance of the structure is observed [1,2,19].

The magnitude of $\Delta h$ multifractality index for the relief profiles of the investigated samples are in the range of 0.5–1.2 (see [2] and Figure 14). Such values are typical of stochastic objects and processes with a strong statistical inhomogeneity observed in nature [20,24]. The multifractality is related with the property of generalized scale invariance in the range from nanoscale to mesoscale. The concept of multifractality is used in the physics of disordered media (for example, quantum phase transitions), developed turbulence of hydrodynamic flows, etc.

The findings of $D(h)$ and dimension indexes in Table 1 have to be used for comparative analysis with other multifractal objects in nature. In mathematics, a multifractal object is described by a cascading mechanism to form a hierarchy of scales. In physics, cascade models are considered, starting with the works of A. N. Kolmogorov on turbulence [21].

The fractal growth can be treated with nonlinear equations (e.g., Kardar–Parisi–Zhang equation [25]). Inhomogeneous stochastic clustering is considered by kinetic models using the Smoluchowski equation [26,27]. It considers two particles (or clusters) with masses $m_1$ and $m_2$ that interact and form a new particle (cluster) with mass $m = m_1 + m_2$, an aggregation is irreversible. The temporal evolution of concentration $N(m,t)$ is governed by the following equation (see [27]):

$$\frac{\partial N(m,t)}{\partial t} = \frac{\Lambda}{2} \int_0^\infty dm_1 dm_2 K(m_1,m_2) \delta(m - m_1 - m_2) - \delta(m - m_1) - \delta(m - m_2)$$

$$+ \frac{\mu}{m_0} \delta(m - m_0) - \frac{1}{2} \delta(m - M)$$

(1)

The kernel $K(m_1,m_2)$ and the factor $\Lambda$ regulate the rate of interaction of clusters (particles). The source of incoming particles of mass $m_0$ with the flux $J_0$ and the sink of particles of mass $M$ with a flux $f$ are considered.

The kernels with self-similarity properties are considered (see [27]):

$$K(hm_1,hm_2,hm) = h^\mu K(m_1,m_2,m), \quad K(m_1,m_2) = m_1^h m_2^v, \quad \mu + v = \eta.$$  

(2)

Indexes $\mu$ and $\nu$ relate with Hurst exponents, which were found from our experiments, Table 1.

A formal analogy between the equations for fragmentation–aggregation and 3-wave turbulence are discussed elsewhere in the scientific literature (see, e.g., [28]). The Kolmogorov–Zakharov approach [21,29,30] is useful to treat the problem. To describe agglomeration the turbulence theory considered cascade models [20,29] can be used. Kolmogorov’s theory [20,21] can be used to treat the kinetic equation with kernel (2).
To simplify the problem, experimental self-similarity indexes describing stochastic surfaces can be used. Scaling exponents and fractal dimensions in Table 1 observed in real clustering processes should be used. Scalings and fractal dimensions (Table 1) will help to describe the dilatation symmetries (scale invariance) of solid materials and agglomerates. Fractal growth is associated with a universal cascade mechanism for the fracture centers formation under nonequilibrium condition, when acoustic unloading of the irradiated object does not provide relaxation, see, e.g., [1,3,25].

The findings of the irradiated material structure deviated from the trivial stochastic granularity (the Brownian surface) open a way to construct materials with high porosity. Such materials with stochastic nano- and microstructure is proposed to control plasma turbulence and to regulate aerodynamics [31]. Future research directions will be developing the technology to regulate stochastic clustering of various symmetry by controlling near-wall plasma turbulence.

6. Conclusions

Materials of initial crystalline structure (titanium, tungsten, carbon, lithium, and beryllium) were analyzed after plasma irradiation in fusion devices. Exceptional extreme high-temperature plasma load in fusion devices leads to a specific surface clustering. It is strictly different from any other conditions of material’s clustering. The hierarchical granularity with cauliflower-like shape and surface self-similarity have been observed. Height’s distribution is deviated from the Gaussian function. The relief roughness differs qualitatively from the ordinary Brownian surface and from clustering under other conditions. The industrial steel casting with the ordinary roughness formed at solidification after melting and the molybdenum sample irradiated in the magnetron plasma were used as reference samples to demonstrate a difference with samples irradiated with plasma in fusion devices.

For samples from fusion devices, the quantitative characteristics of materials’ structure are Hurst exponents from 0.55 to 0.9. The fractal dimension of the surface from 2.1 to 2.45 was estimated. Non-homogeneous stochastic clustering is described by the multifractal spectrum broadening 0.5–1.2. It is in the range observed for typical multifractal objects in nature.

In fusion devices, the specific conditions regulate material surface clustering faced to plasma. Ions and clusters on the surface move under the effect of fluctuated electric field driven by the near-wall turbulent plasma. In such a process, long-term correlations lead to the growth of surface with a self-similar structure. The multi-scale effects influence the self-similarity–fractal growth from nanometers to millimeters. Collective and synergistic effects dominate in such stochastic clustering. Experimental results illustrate universal process of stochastic clustering of materials irradiated with the high-temperature plasma in fusion devices.

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