A comparative study of correlation methods for determination of fractal parameters in surface characterization

Slawomir Kulesza [a],*, Miroslaw Bramowicz [b]

[a] Faculty of Mathematics and Computer Science, University of Warmia and Mazury, Sloneczna 54, 10-710 Olsztyn, Poland
[b] Faculty of Technical Sciences, University of Warmia and Mazury, Oczapowskiego 11, 10-719 Olsztyn, Poland

ARTICLE INFO

ABSTRACT

An analysis of several methods of extraction of fractal parameters from the simulated, artificial surfaces and AFM images of the real, polycrystalline diamond films is presented in the paper. The methods involve the cube count method, the roughness method, the autocorrelation function method, and the structure function method. By comparing the four methods, the roughness method is found to be superior for its high numerical accuracy, whereas the cube count method appears to be inferior in that aspect. The changes in the fractal dimension and the anisotropy ratio values observed over deposition time are also shown and discussed in the paper.

© 2014 Elsevier B.V. All rights reserved.

1. Introduction

Precise description and control of 3D surface topography is of prime importance in engineering applications as it has strong influence on such materials properties as: fracture toughness, wear resistance, lubrication and others. Currently, surface characterization basically relies on a large series of statistical parameters derived using various methods, among which atomic force microscopy (AFM) now became one of the most popular. Since its discovery, AFM significantly evolved toward characterization of surface topography of solids of all types down to the nanoscale level, and nowadays this non-destructive method provides topographical information probed over an area from several square nanometers up to thousands of square micrometers. Moreover, this method can discriminate among a large diversity of interactions occurring between the surface and the scanning tip (electrical, magnetic, adhesive, friction, etc.)

Surface topography can be characterized using an excessive number of statistical parameters depicting various aspects of the surface lay, roughness, waviness and the form. Unfortunately, many of them strongly depend on how they are actually measured, including for instance the sampling and the scan lengths, and the instrumental resolution. In order to overcome this problem, description of engineering surfaces in terms of fractal geometry was suggested [1]. Fractals are virtual, self-similar geometrical

objects that appear identical independent of the scale of magnification. Such objects are characterized by the fractal dimension D [1]. However, due to some physical confinements (finite instrumental resolution, finite observation time), fractal properties of objects of natural origin are often reduced to a limited range of scale lengths. Such objects (called self-affine) need to be described by three parameters: fractal dimension D, corner frequency f_c, and the topothesy Λ, and differ from perfect fractals described by the fractal dimension D solely.

Fractal dimension is found to be correlated with surface roughness parameters [2–5], it is related to various material properties [6], and even to mechanisms leading to surface formation [7]. On the other hand, several experimental methods have been proposed for estimation of fractal dimension, including for example: AFM, Scanning Electron Microscopy [8], diffuse X-ray reflectometry [8], adsorption measurements, electrochemical impedance spectroscopy and others, which usually hardly converged into a consistent picture. What is worse, a little is known about possible influence of each numerical procedure on final results. In previous work we have compared the effect of the AFM tip geometry and the scan mode on results of a fractal analysis of well-established surfaces (calibration gratings) [4]. In this study we report the systematic comparison of different numerical procedures used to estimate the fractal dimension from the same AFM images recorded from the crystal surface that evolves with elapsed time. Such an attempt should exhibit numerical peculiarities allowing the quantitative assessment of processing procedures, which might be especially useful in fulfilling lacking knowledge about their accuracy, precision and terms of applicability.

* Corresponding author. Tel.: +48 89 524 6075; fax: +48 89 524 6089.
E-mail addresses: kulesza@matman.uwm.edu.pl (S. Kulesza),
mbramowicz@moskit.uwm.edu.pl (M. Bramowicz).

The selection of thin diamond films grown by microwave plasma was motivated by several reasons. First, the deposition process significantly affects the crystal morphology that starts from a bare substrate surface with tiny nuclei, and evolves into a closed although polycrystalline film with increasing roughness, which is expected to influence the fractal properties. In addition, a little is known about fractal properties of diamonds itself. Salvadori et al. [9] reported D value close to 2.5 for plasma-deposited diamond films on silicon substrates, whereas Silva et al. [10] obtained lower D equal to 2.2 for boron-doped diamond films grown by the hot-filament CVD method. Recently, Tsysar [11] found fractal dimension equal to 2.36, and 2.73 for [111]- and [011]-oriented diamond grains in HF-CVD films, respectively. Finally, a modelization of the topography evolution in terms of fractal parameters might shed some light onto the kinetics of the growth process, including the early stages of the nucleation phenomena, and subsequent diffusion of contaminants and defects along grain boundaries.

2. Materials and methods

Diamond films were deposited on 1 mm thick substrates made of fused quartz. Pre-treatment procedure involved mechanical seeding with 250 nm diamond powder on a vibrating plate. The growth process was carried out in a microwave plasma CVD reactor (ASTeX AX 6560) described in details elsewhere [12]. Gas mixture contained methane largely diluted with molecular hydrogen ($CH_4/(CH_4 + H_2) = 5\%$ (vol.)) Other deposition parameters were as follows: substrate temperature $500\,^{\circ}C$, gas pressure 6650 Pa (50 Torr), microwave power 3000 W, and deposition time varying from 5 min up to 5 h.

AFM measurements were carried out at ambient conditions using Multimode 8 instrument with Nanoscope V controller (Bruker). The tip (SNL-10 (Bruker)) with the radius 2 nm scanned across the surface in a contact mode. To determine the fractal properties, square AFM images with the lateral resolution of 512 points and scan lengths from 1 up to 150 μm were taken. The images were then flattened to remove line tilt and image bow prior to further numerical processing.

3. Evaluation of the fractal dimension

3.1. The cube counting method

An evaluation of the fractal dimension by counting the cubes directly explores the definition of a box-count dimension. The algorithm iteratively halves an initial cubic cell with the edge length L equal to the scan length into smaller cubes, and counts $N(L)$ – the number of all cubes that contain at least one sample of a 3D topography. The process continues until L approaches the image resolution, i.e. the distance between two adjacent samples [13]. Since:

$$N(L) \propto L^{-D} \tag{1}$$

the slope of a log–log plot of $N(L)$ versus L gives the fractal dimension referred to as the cube count fractal dimension D_{CC}.

3.2. The roughness method

The fractal dimension can be also estimated using the root-mean-squared value of the surface height variance S_q defined as [14,15]:

$$S_q = \sqrt{\frac{1}{N_x N_y} \sum_{i=0}^{Nx-1} \sum_{j=0}^{Ny-1} (z(i,j) - \langle z \rangle)^2} \tag{2}$$

where $<\cdots>$ – denotes mean value, N_x, N_y – is the number of samples along rows and columns in the AFM image, while $z(i,j)$ is the measured height in pixel (i,j) of an image. Assuming that the roughness S_q measured over surfaces with different edge lengths L scales as:

$$S_q \propto L^{3-D} \tag{3}$$

the fractal dimension D_{RMS} can be computed from the slope of a least-square regression line fit in a log–log plot of S_q vs. L [16].

3.3. The structure function method

The surface topography recorded in the form of discrete height samples $z(i,j)$ in an AFM image allows us to compute the three-dimensional structure function (SF) defined as:

$$S(\tau_x, \tau_y) = \left\langle (z(x, y) - z(x + \tau_x, y + \tau_y))^2 \right\rangle ;$$

$$\tau_x = \frac{L_x}{N_x} \quad (0, 1 \ldots N_x - 1), \quad \tau_y = \frac{L_y}{N_y} \quad (0, 1 \ldots N_y - 1) \tag{4}$$

where $<\cdots>$ – denotes the spatial average, N_x, N_y – are the numbers of samples along scan axes, L_x, L_y – scan lengths, whereas (τ_x, τ_y) – the discrete spatial lag along scan axes between an original image and its delayed copy. Any profile of the structure function derived from the image is assumed to obey the approximate scaling-law behavior:

$$S(\tau) = \Lambda \tau^{2(2-D)} \tag{5}$$

where D – is the profile fractal dimension, while Λ – is the topothesy. According to Wu [17], the topothesy can be expressed explicitly as:

$$\Lambda = \frac{\pi G^{2(D-1)}}{2\Gamma(5 - 2D)\sin[\pi(2 - D)]} \tag{6}$$

where G – is a scale constant with the dimension of reciprocal length, D – is the profile fractal dimension ($1 < D < 2$), and Γ – is the Euler function.

Note that any section through the structure function at an arbitrary angle around the origin would be equivalent to an ensemble average of profile structure functions measured at the angle Θ_τ with respect to the x-axis:

$$\Theta_\tau = \tan^{-1}\left(\frac{\tau_y}{\tau_x}\right) \tag{7}$$

The profile fractal dimension D is then calculated from the least-square regression line in a log–log plot of the profile structure function versus separation lag for each Θ_τ angle, and the surface fractal dimension D_{SF} is computed according to:

$$D_{SF} = \langle D \rangle + 1 \tag{8}$$

where $<\cdots>$ denotes tangentially averaged mean fractal dimension. Likewise, the topothesy Λ can be determined from the intercept of the above plot with the y-axis.

3.4. The autocorrelation function method

According to Nayak [18], any surface represents a real random process with the spatial variation described by the autocorrelation function (ACF). Assuming stationarity and ergodicity, the ACF can be computed through spatial averaging over a limited number of AFM samples. The stationarity condition requires that both the autocorrelation function and the mean value are independent of the position, whereas the ergodicity requires the mean to converge to a constant value with increasing sampling period. Even surfaces with apparent curvature and waveform (i.e. non-stationary) can be