Fractal parameterization analysis of ferroelectric domain structure evolution induced by electron beam irradiation

A G Maslovskaya and T K Barabash
Physics Engineering Department, Amur State University, 21 Ignatyevskoe Shosse, Blagoveshchensk 675000, Russia
E-mail: maslovskayaag@mail.ru

Abstract. The article presents some results of fractal analysis of ferroelectric domain structure images visualized with scanning electron microscope (SEM) techniques. The fractal and multifractal characteristics were estimated to demonstrate self-similar organization of ferroelectric domain structure registered with static and dynamic contrast modes of SEM. Fractal methods as sensitive analytical tools were used to indicate degree of domain structure and domain boundary imperfections. The electron irradiation-induced erosion effect of ferroelectric domain boundaries in electron beam-stimulated polarization current mode of SEM is characterized by considerable raising of fractal dimension. For dynamic contrast mode of SEM there was revealed that complication of domain structure during its dynamics is specified by increase in fractal dimension of images and slight raising of boundary fractal dimension.

1. Introduction
Nowadays SEM techniques have become increasingly important in many areas of scientific work and in practice. The unique SEM analytical abilities of registration of specific responses electron beam-induced allow thoroughly exploring materials which possess high sensitivity with respect to external exposure. The urgent applications of SEM consist in surface topography visualization as well as study of potential relief of polar materials [1]. In this way as from middle of the last century SEM capabilities were employed to obtain images of domain structures, to study and to modify electric properties of ferroelectric materials. In this case one of the key problems consists in studying of sample modification which can be potentially stimulated by electron beam irradiation. In the series of studies some unconventional SEM modes have been described to analyze and to modify ferroelectrics. In particular, electron beam polarization current mode [2] enables domain structure to be visualized and simultaneously polarization switching current to be registered.

Notice that fractal geometrical features of ferroelectric domain structure have been previously reported in [3-13]. The formation of fractal ferroelectric domain structures results from self-organization process of physical system. So as to identify self-similar character of ferroelectric domain structure concepts of fractal theory can be used [14-15]. The fractal analysis yields one of the directions of mathematical description of wide class of objects which have complex and nonregular structures. The fractal analysis of geometry of ferroelectric domain structure images obtained with SEM can be characterized by specific features because of potential contrast pattern in SEM is due to electron beam interaction with samples. Using secondary electrons emitted from surfaces of domains with inverse polarizations allows us to register potential contrast of a specimen. Videosignal and as a consequence observed contrast indicate polarization processes in ferroelectrics in some active probing
modes specifically in electron beam induced polarization current mode [2]. Therefore inhomogeneities of the images correspond to variation of polarization distribution. This can be attributed to accumulation injected charge, distributions of defects and stochastic nucleation process (Barkhausen effect). The fractal analysis has become sensitive technique to study of domain boundary irregularity and it can be used for quantitative characterization of observed modification of domain structures. The present study is therefore aimed to analysis of fractal characteristics of ferroelectric domain structure images visualized with static and dynamic modes of the SEM.

2. Research methodology outline
First in order to estimate the scaling characteristics of SEM-images we can apply the algorithms of fractal and multifractal analysis designed for two-dimensional raster images. The fractal dimension of object is a major quantity of fractal theory. The fractal dimension yields numerical estimation of inhomogeneity and nonregularity of self-similar structure. Here covering method based on ratio of “number of particles $N$ – measure” can be applied [14]:

$$N = (\delta / \delta_0)^D,$$

where $\delta$ or $\delta_0$ – variable covering parameters, $D$ – fractal dimension of the object.

The equation (1) enables one to obtain the technique for calculation of fractal dimension $D$ of physical object providing a variation of one of characteristics $\delta$ or $\delta_0$. In particular, box-counting method assumes a variation of the sizes of particles of the covering set $\delta_0$. A binary image is divided into $2^n$ parts covering the raster image. Then the number of element $N$ is counted which contains even if the one point of the image. After that relation between number of element $N$ covering the analyzed image and linear size of element $\delta_0$ is defined both in logarithmic scale using the following equation [14]:

$$\ln N = -D \cdot \ln \delta_0.$$  

As a consequence the fractal dimension $D$ can be calculated from the approximation of the function $N(\delta_0)$ by means of least square procedure.

In order to estimate the fractal dimensions of cluster structures we can use the dependence between perimeter $P$ and area $S$ of a cluster provided clusters or “islands” can be assigned to image [15]:

$$P = C \cdot \delta^{1-D} \cdot S(\delta)^{D/2}.$$  

Assuming that the unit is equal to measure of length $\delta$ (it is the one pixel for a raster image). In this case the relation between perimeter and square (3) can be specified with the following equation:

$$\ln P = (D/2) \cdot \ln S.$$  

Whereas complexity and irregularity of analyzed domain structures can take place the authoring modification of algorithm was designed to calculate fractal dimension of bitmapped images of domain boundaries. Modification is based on equation (1) and the following principles are assumed. The process of cluster marking is realized by means of cluster identification Hoshen-Kopelman algorithm [16] with the recursive procedure excluding incorrect cluster marking in case of repeated viewing.

Moreover the method of multifractal parametrization [14-15] and the method of wavelet transformation modulus maxima [17] were also applied to analyze more complex images of domain structures which can be characterized as self-affine objects. These methods permit one to estimate multifractal scaling properties considering in details some images which representing nonuniform fractal objects. Multifractals due to having statistical geometrical properties require fractal dimensions spectrum to describe them. Compared with fractal analysis, the initial image is divided into clusters with the subsequent reduce in cluster size $l$. Then a matrix $C$ is created provided each element of $C$ is equal to number of the filled cells. In this case image consisting of $N \times N$ points has in a $(i,j)$ cluster $(i, j = 1, k, k = N/l)$ a determined quantity of points. In multifractal parametrization not only the quantity of the clusters comprising points of self-similar structure is counted, but also their specific weight [15]:
where $\sum C_{i,j}$ is total number of units in a matrix of clusters.

Then we use the measure $M_d(q,l) = \sum_{i,j,l,k} P_{i,j}^q \cdot l^d = N(q,l)l^d$, where $l$ is the cell size, $q$ is a moment order. Change in $P_{ij}$ depending on $l$ according to power law is defined by sequence of the factors $\tau(q)$ characterizing a measure. Further the weighed number of cells $N(q,l)$ can be expressed in a form: $N(q,l) = \sum_{i,j,l,k} P_{i,j}^q \propto l^{-\tau(q)}$ and the indicator of weight is defined by formula $\tau(q) = -\lim_{l \rightarrow 0} (\ln N(q,l)/\ln l)$.

Therefore calculation of the $N(q,l)$ and $\tau(q)$ is performed at a variation of parameter of deformation of $q$. The Renyi’s multifractal spectrum can be evaluated by the equation

$$D(q) = \tau(q)/(q-1).$$

Algorithmization and program implementation of examined methods were performed to study scaling characteristics of SEM domain structure images. All of the initial images of ferroelectric domain structures were initialized as raster images represented in a bitmap format.

### 3. Application of fractal analysis methods to ferroelectric domain structure images obtained with static contrast mode of the SEM

Previous studies have indicated that analysis of geometrical properties of ferroelectric domain structure images visualized with SEM techniques possesses a number of specific features owing to the fact that the potential contrast image is result of electron beam interaction with the sample. In this terms tonality of domains as well as domain boundaries observed with SEM is defined by features of the used mode as well as mechanisms of contrast formation. In particular, at first the electron probe initiates specific effects in the irradiated sample, and subsequently registers them in the “active” scanning modes. One of priority factors of fractal analysis of ferroelectric domain structures consists in correct interpreting the images of the analyzed element of domain structures and correct submitting initial data.

The present study confirms that SEM images of static domain configurations are characterized by a complex scaling. The fractal dimensions of ferroelectric domain structure images were calculated by means of box-counting method (2). The results suggest fractality of images and distribution of fractal dimensions over the range from 1.6 to 1.9 for typical ferroelectrics. The fractal dimensions were evaluated for set of images of each objects. Whence we obtained the following results: $D=1.72–1.88$ for Rochelle salt, $D=1.71–1.92$ for triglycine sulfate (TGS), $D=1.60–1.77$ for barium titanate (BaTiO$_3$), $D=1.84–1.89$ for bismuth titanate (Bi$_2$Ti$_3$O$_7$), $D=1.64–1.78$ for lead titanate (PbTiO$_3$), $D=1.86–1.95$ for sodium nitrite (NaNO$_2$), $D=1.52–1.56$ for PZT ferroelectric ceramics. However the classical box-counting method enables us to estimate only the quantitative characteristics of space filling of fractal and it does not allow describing form of structures of which the image consists.

The irregularity and roughness degree of domain boundaries for typical ferroelectrics was identified by slit island method (4) and it is characterized by fractal dimension from the almost regular forms with $D=1.05$ to strongly curved with pronounced fractal properties ($D=1.66$). For instance, the fractal dimension of the domain boundaries separating $90^\circ \ a$-components of spontaneous polarization and boundaries separating antiparallel $180^\circ \ c$-components in monocrysals of Bi$_4$Ti$_3$O$_{12}$ was the quite considerable and it is distributed over the range of $D=1.509–1.780$. The example of result of fractal dimension calculation for the line restricting clusters ($a$- and $c$-domains) is shown in figure 1 a.

The performed multifractal analysis for domain structure images has allowed us to calculate the spectral characteristics specifying dispersion measure of fractal dimensions. The figure 1 b illustrates
the Renyi’s multifractal spectrum (6) computed for typical domain structure of TGS crystal (positive domains or dark contrast were subject to the analysis).

![Graph](image)

**Figure 1.** The result of fractal analysis performed by slit island method for bismuth titanate domain structure image shown in the insert figure – a; the Renyi’s multifractal spectrum for TGS crystal – b.

The computed scaling factor \( \tau(q) \) is nonlinear which is relative to multifractal structures. The analysis of Renyi’s multifractal spectrum permits us to define a set of the dimensions forming the structure and also to estimate width of a fractal spectrum (\( \Delta \alpha \approx 2 \)). The value of \( D(q=0)=1.86 \) corresponds to the fractal dimension calculated by covering method. The analyzed image of TGS domain structure has a self-similar structure with distribution of fractal dimension over the range of 1.7 – 3.7. The wavelet transformation modulus maxima method allows visualizing a picture of wavelet-coefficients and extremum skeleton which show hierarchical structure of a profile of the image. Also this method leads to the spectral characteristics of \( \pi(q) \) and \( f(q) \) which are in good agreement with the data obtained by multifractal parametrization method.

The fractal analysis of image of lens-like domain boundary of TGS crystal observed with electron beam induced polarization current mode [2] at probe current \( I=10^{-9} \) A and beam accelerating voltages \( U=3-5 \) kV so long as irradiation time was varied. Microphotographs and corresponding fractal dimensions calculated by slit island method (4) are demonstrated in figure 2.

![Microphotographs](image)

**Figure 2.** The TGS domain boundaries observed with electron beam stimulated polarization current mode and corresponding values of computed fractal dimensions: a – the image of boundary of lens-shaped domain, b – erosion of boundary under prolonged irradiation, c – the image of domain fragment after 5 minute of irradiation.

The fractal dimensions were estimated to express roughness and irregularity domain degree. The durational irradiation of sample leads to erosion of image: line of boundary disintegrates on series of the points and becomes wider as it is shown in figure 1 c. The erosion could be caused by modification of boundary area structure by means of creating ramified antiparallel structure with width up to 10 \( \mu \)m. Note also that increase in fractal dimension indicates complication of ferroelectric domain geometry.

4. Estimation of fractal characteristics of ferroelectric domain structure images visualized with dynamic contrast mode of the SEM

Taking into consideration performance capabilities of dynamic contrast imaging modes, there is a practical interest examining evolution of fractal dimension during sequential stages of domain...
structure dynamics induced by an electron beam of SEM. Let us perform analysis of experimental data obtained with potential electron contact mode [2].

The fractal dimensions of sequential stages of domain structure dynamics in barium titanate (BaTiO$_3$) were calculated by box-counting method (2). The result can be attributed to complication of domain structure with fractal dimension distribution of the following range: $D=[1.608,1.613,1.729,1.734]$. Meanwhile, the degree of irregularity and roughness of domain boundaries changes slightly: $D^*= [1.261,1.263,1.269,1.277]$. Providing Bi$_4$Ti$_3$O$_{12}$ is subjected to field directed perpendicular to the sample surface in potential electron contact mode, we can conclude that electron beam exposure results in movement of boundaries dividing anti-parallel $c$-components of spontaneous polarization.

The domain growth with parallel to field $c$-component of spontaneous polarization is realized by expansion of the wedges which are formed on 90° domain wall. This process is specified by increase in the fractal dimension from $D=1.784$ to $D=1.807$ which is calculated by covering method. On the other hand, fractality of domain boundaries remains in rather narrow interval $D \in (1.61,1.66)$. Characteristic feature of polarization switching process in TGS is reproducibility of process of time cycle. The analysis of the scaling characteristics of SEM-contrast of TGS domains during domain structure dynamics observed in the potential contact mode was carried out using of the videofile with visualization of a full cycle of polarization switching in TGS [2], which is illustrated in figure 3. In order to obtain data suitable for the fractal analysis quantization of video with an interval of 1 s has been performed.

![Figure 3](image_url)

**Figure 3.** Fragments of SEM-images of TGS domain structure during complete cycle of polarization reorientation in potential electron contact mode [2] characterized by fractal dimension distribution over the range of (1.012, 1.214).

The fractal analysis of domain boundary geometry with use of slit island method (4) yields change in dimension in the interval $D \in (1.012,1.214)$ with a growth of both positive and negative domains. The minimum value of fractal dimension corresponds to the initial stage of domain nucleation with practically regular boundaries. The maximum value is attributed to critical state: the square of the switched area is equal to the square of nonswitched area. To analyse the fractal dimension of images computed by box-counting method (2) the temporal dependence of fractal dimension is represented in figure 4 a. These data can be acceptable approximated by quadratic function.

![Figure 4](image_url)

**Figure 4.** The temporal dependence of fractal dimension calculated by box-counting method – $a$, the dynamics of changes in fractal dimension for positive (o) and negative (x) domains – $b$. 
Providing we consider temporal dependence of fractal dimension for growth of positive and negative domains on the one graphic we can conclude that complication of domain structure for growth of both positive and negative domains occurs almost identically as shown in figure 4 b. The evidence from this study suggests that visually observed features of domain structure dynamics can be attributed to peculiarities of change in fractal dimension. In general, therefore, it seems that not only static and quasi-static domain configurations are characterized by a fractal structure but also domain dynamics induced by an electron probe demonstrates self-organization process.

5. Concluding remarks
The most obvious finding to emerge from this study is that fractal and multifractal methods are sensitive techniques to study degree of domain structure and domain boundary imperfections and can be used for the quantitative characteristic of observed modifications of ferroelectrics in SEM, in particular, are caused by accumulation of the injected charge, defect distribution, and stochastic nucleation process (Barkhausen effect).

Our data suggest electron beam-induced erosion effect of domain boundaries in TGS can be characterized by increase in fractal dimension from \( D = 1.245 \) to \( D = 1.521 \). Also complication of domain structure geometry during its dynamics is characterized by increase in value of fractal dimension of the image and insignificant increase in fractal dimension of boundaries when analyzing ferroelectrics with dynamic contrast modes of SEM.

The evidence from fractal analysis for step-by-step images of a full cycle of polarization switching in TGS in the potential electron contact mode indicates that complication of domain structure for the lateral growth of positive, as well as negative domains occurs similarly: fractal dimension of the image changes within the interval \( 1.63 < D < 1.99 \) and domain boundaries \( 1.012 < D < 1.214 \) respectively.

Acknowledgments
The work has been supported by a scientific program of the Ministry of Education and Science of Russia (project No. 1158.2014/9).

References
[1] Robinson G Y and White R M 1967 Appl. Phys. Lett. 10 320
[2] Sogr A A, Maslovskaya A G and Kopylova I B 2006 Ferroelectrics 341 29
[3] Ozaki T, Fujii K and Ohgami J 1995 J. Phys. Soc. Jpn. 64 2282
[4] Isupov VA 1998 Physics of the Solid State 40 1188
[5] Jeng Y-R, Tsai P-C and Fang T-H 2003 Microelectronic Engineering 65 406
[6] Shur V Ya, Kuznetsov D K, Lobov A I et al 2006 Ferroelectrics 341 85
[7] Wu Z, Duan W, Wu J and Gu B-L 2007 Nanotechnology 18 325703
[8] Roy M K, Paul J and Dattagupta S 2010 IEEE Xplore 109 014108
[9] Galiyarova N M, Bey A B, Kuznetsov E A and Korchnariyuk Y I 2004 Ferroelectrics 30 205
[10] Tadic B 2002 Eur. Phys. J. B. 28 81
[11] Maslovskaya A G and Barabash T K 2012 Physics Procedia 23 81
[12] Maslovskaya A G and Barabash T K 2013 Ferroelectrics 442 18
[13] Pavelchuk A, Barabash T and Maslovskaya A G 2016 IOP Conf. Series: Materials Science and Engineering, 110 012080
[14] Bojokin S V and Parshin D A 2001 Fractals and multifractals (Moscow: R&C Dynamics)
[15] Falconer K 1990 Fractal Geometry: Mathematical Foundations and Applications (Chichester: Wiley)
[16] Stauffer D and Aharony A 1992 Introduction to Percolation Theory (London: Taylor & Francis)
[17] Arneodo A, Decoster N and Roux S G 2000 Eur. Phys. J. B. 15 567