

## ENTROPY AND FRACTAL NATURE

by

**Vojislav V. MITIĆ<sup>a,b\*</sup>, Goran M. LAZOVIĆ<sup>c</sup>, Jelena Ž. MANOJLOVIĆ<sup>d</sup>,  
Wen-Chieh HUANG<sup>e</sup>, Mladen M. STOJILJKOVIĆ<sup>d</sup>,  
Hans FACHT<sup>f</sup>, and Branislav VLAHOVIĆ<sup>g</sup>**

<sup>a</sup> Faculty of Electronic Engineering, University of Nis, Nis, Serbia

<sup>b</sup> Institute of Technical Sciences of SASA, Belgrade, Serbia

<sup>c</sup> Faculty of Mechanical Engineering, University of Belgrade, Belgrade, Serbia

<sup>d</sup> Faculty of Mechanical Engineering, University of Nis, Nis, Serbia

<sup>e</sup> University Department of Chemistry and Frontier Research Center on Fundamental and Applied Sciences of Matters, National Tsing Hua University, Hsinchu City, Taiwan

<sup>f</sup> Institute of Functional Nanosystems, University of Ulm, Ulm, Germany

<sup>g</sup> North Carolina Central University, Durhan, N. C., USA

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*Existing, the biunivocal correspondents between the fractal nature and the nature discovered by fractals is the source and meeting point from those two aspects which are similar to the thermodynamically philosophical point of view. Sometimes we can begin from the end. We are substantial part of such fractals space nature. The mathematics fractal structures world have been inspired from nature and Euclidian geometry imagined shapes, and now it is coming back to nature serving it. All our analysis are based on several experimental results. The substance of the question regarding entropy and fractals could be analyzed on different ceramics and materials in general. We have reported the results based on consolidation BaTiO<sub>3</sub>-ceramics by the standard sintering technology, performed with BaTiO<sub>3</sub> and different additives (MnCO<sub>3</sub>, CeO<sub>2</sub>, Bi<sub>2</sub>O<sub>3</sub>, Fe<sub>2</sub>O<sub>3</sub>, CaZrO<sub>3</sub>, Nb<sub>2</sub>O<sub>5</sub>, Er<sub>2</sub>O<sub>3</sub>, Yt<sub>2</sub>O<sub>3</sub>, Ho<sub>2</sub>O<sub>3</sub>). Thermodynamic principles are very important. Beside the energy and temperature, the entropy as a measure between the order and disorder (chaos) is very important parameter. In this paper, we establish the relation between the entropy and fractal that opens new frontiers with the goal to understand and establish the order-disorder relation.*

Key words: *ceramics materials, thermodynamics, entropy, fractals, order-disorder*

### Introduction

We have developed numerous methods for micro-structure modeling grain geometry, grain boundary surface and contacts, what is of the basic importance for electric and other properties optimization. Most of these methods are based on contact surfaces calculations in the prescribed volume of the ceramic sample, and on defining shape modeling. The Euclidian geometry characterization has been performed in contact surfaces calculation, at the very beginning of all of these morphological structure analyses.

\* Corresponding author, e-mail: vmitic.d2480@gmail.com

The fractals world is everywhere, all around us. There is self-similarity which, in itself implies recursion, makes them suitable for processing in applications from the disorder to the order. Anyhow, it is very important to have them as a measure of self-identity and that would uniformly provide support for biunivocal penetration. The property of physical objects shares some features characteristic to fractal objects, and they are the crucial concept of modern scientific view on the World, tightly connected with the fractal nature view process of the order-disorder relation. The trend in the recent results, is that a wide range of disordered systems, *e. g.*, linear and branched polymers, biopolymers and percolation clusters can be characterized by the fractal nature over a microscopic correlation length [1].

There is very specific relation within the matter reality, which was always treated as a thermodynamical system. Thermodynamic functions and fundamental parameters such as temperature, Gibbs free energy, enthalpy and entropy are very important to understand the processes in materials world. We understand the entropy as a measure of order within the system in disorder. There is the great philosophical and scientific interest to control chaotically process and structures. In that way we could try practically to control the border between chaos and order. It is a great challenge to find appropriate approach for this open question and on that we can make amazing contribution to worlds existing secrets in modern science.

### Short introduction on fractals

The most important term in theory of fractals was established by Mandelbrot and Frame [2] and Mandelbrot [3]. Explanation of the role of fractal dimension will be described below.

We can understand that the more irregular the stone has higher fractal dimension. Hausdorff dimension or fractal dimension  $DH_f$  is real number to the contrast of usual notion of dimension which is called topological dimension  $DT$  ( $DT = 0$  for isolated points,  $DT = 1$  for curves,  $DT = 2$  for surfaces,  $DT = 3$  for solids, *etc.*). So, Hausdorff (or fractal) dimension  $0 < DH_f < 1$  covers all objects that are more than a point but less than a curve. Good example is Cantor set with  $DH_f = \ln 2 / \ln 3 \approx 0.6309$ .

Cantor set is mathematical construction. Direct analogy to Cantor set is the dust that floats in the air. Brian Kaye devoted fractality of different dusts: mine dust, fumed silica, radioactive dust, and powders for ceramics industry [4-6].

If  $1 < DH_f < 2$ , the object is something between curve and surface. Good examples are coastlines. The coastline of Ireland has a fractal dimension approximately 1.22 while of Great Britain is 1.25 and of Norway is the most irregular with  $DH_f \approx 1.52$ .

The maritime coasts are just level lines of geomorphic relief formations, and the same approach can be applied to ceramics grains. Therefore, there is a reason to study grains contours whose lines are more or less complicated.  $BaTiO_3$  grains as 3-D objects have fractal surfaces, which means that their fractal dimension is the real number between 2 and 3. Theory shows that any surface generated by regular Brownian motion has  $DH_f = 2.5$  which is the same as fractal dimension of crumpled paper.

Let  $M$  be measure of the object  $S$  characterized by length, area, volume or hyper-volume, but also mass, charge of electricity, *etc.* If the side length of 3-D cube doubles, then  $M(2l) = 2^3 M(l)$ . If the side triples, then  $M(3l) = 3^3 M(l)$ , fig. 1.

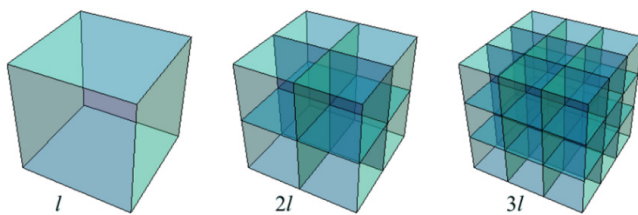


Figure 1. Cube side length multiplication

In general:

$$M(\lambda l) = \lambda^{DH_f} M(l) \quad (1)$$

This functional equation has nontrivial solution  $M(l) = Cl^{DH_f}$ ,  $DH_f$  is fractal dimension of set S.

The BaTiO<sub>3</sub> grains surfaces fractal dimensions are slightly above surface's topological dimension  $D_T = 2$ . The difference  $DH_f - D_T = DH_f - 2$ , is thereby supposed to be responsible for affection to the part of ferroelectric phenomena in this ceramics, that cannot be explained by purely grain surfaces Euclidean geometry. It is suitable to introduce a normalized surface fractality parameter  $\alpha_S$  satisfying the inequality:

$$(1 - \varphi) \min\{DH_f - 2\} < \alpha_S < \varphi \max\{DH_f - 2\}, \quad 0 < \varphi < 1 \quad (2)$$

which ensures the unit range  $0 < \alpha_S < 1$ .

The BaTiO<sub>3</sub> ceramics as a porous material corresponds to a lacunar fractal models. It brings in a new phenomenon, solidification of porous and *spongy* materials increases overall fractal dimension from 2 to full solid 3. In other words, fractal dimension of a porous material,  $DH_p$  satisfies  $2 < DH_p < 3$ . It causes another correction factor  $\alpha_p = D_T - DH_p$ , where  $D_T = 3$  is dimension of the space and  $DH_p$  is corresponding fractal dimension of a porous configuration. Therefore:

$$0 < \alpha_p < 1 \quad (3)$$

The dimensionless quantities  $\alpha_S$  and  $\alpha_p$  will be called geometric fractality factors. We suggest the existence of the third factor  $\alpha_M$  caused by motion disorder influence of ferroelectric particles that is factor of fractal motions.

It is known, that there is a moving *cloud* of moving particles in semiconductors consists of electrons in atoms with large atomic numbers, nucleons in heavy atomic nuclei, and gases consisting of quasi particles with half-integral spin, called Fermi gas.

So, the Fermi gas real dynamics impose necessity of inclusion the fractal motions factor  $\alpha_M$ , that makes third fractality factor, beside  $\alpha_S$  and  $\alpha_p$ . Since, Fermi gas particles have dynamics similar to 3-D Brownian one,  $\alpha_M$  should be derivate of Brownian 3-D space-filling curve Hausdorff fractal dimension. It is obvious that  $1 \leq DH_f \leq 3$ . By continuity of particle trajectory  $\min DH_f = 1$ . It gives maximum of trajectory complexity in 3-D space. It is reasonable to normalize quantity  $\alpha_M$ , by taking:

$$\alpha_M = \frac{1}{2}(DH_f - 1) \quad (4)$$

which gives  $0 < \alpha_M < 1$ . In this way, three independent dimensionless fractality factors  $\alpha_S$ ,  $\alpha_p$ , and  $\alpha_M$  are introduced. These are real numbers from the open interval (0, 1).

### Experimental part

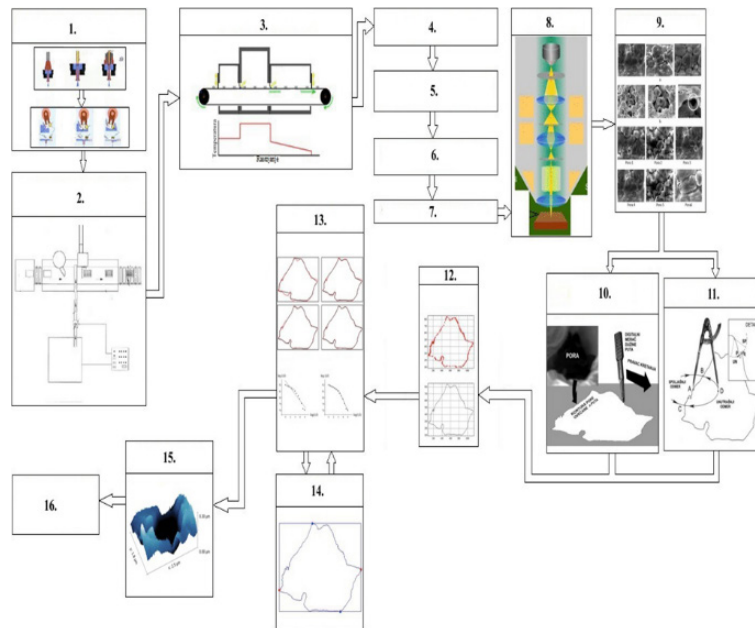
The samples used in our paper were prepared by a conventional solid-state reaction. As the starting powder it was used BaTiO<sub>3</sub> (Murata) doped with different additives in concentration from 0.5 to 5.0 wt.%. Starting powders were ball milled in ethyl alcohol, drying for several hours, and pressed into pellets at a pressure from 90 to 120 MPa. The pellets were sintered in air from 1180 °C to 1380 °C for 2 and 4 hours. The microstructure was investigated by scanning electron microscope, JEOL, SEM-5300 equipped by energy dispersive spectrom-

eter system. The electrical characteristics were measured using LCR meter Agilent 4284A. The micro-structures have been done with selection of some grains and pores with minimum five magnifications, because of fractal nature analysis.

Here, we present the principle scientific-technological-research between different blocks of the schematically and also laboratory process for ceramics consolidation, especially models of the grains and pores reconstructions based on fractal nature analysis which is under development and patenting, tab. 1 and fig. 2.

**Table 1. Details of experimental procedure for samples consolidation and fractal nature micro-structure analysis and grains reconstruction**

1	Powders preparation and samples molding
2,3	Loading the samples and sintering process in furnace
4,5,6	Samples preparation for electrophysical testing
7	Samples preparation for SEM characterization
8	Samples characterization by SEM
9	Micro-structures with minimum five different magnifications per structure
10,11	The grains perimeters measuring by run-meter by compass
12,13	The fractal analysis, perimeters reconstruction and fractal dimension extraction
14	Besicovitch-Minkowski hull, and establishing the grain boundarie double layer contact zone
15	The grains fractal reconstruction
16	New creations and applications in the area of further electronic circuits miniaturizations



**Figure 2. Integrated experimental procedure for fractal nature micro-structure analysis and grains**

### ***Practical procedure***

Analytic method and fractal nature application in reconstruction of materials structure, grains and pores, in order to make an advance prognosis of designed micro-structural

properties is a new procedure in materials micro-structural characterization. Electronic microscopy methods, regardless on resolution and magnification, lead only towards getting the microscopic pictures (micro-graphs). Based on the grains and pores perimeters fractal analysis block (10, 11), their reconstruction is made block (12), and more realistic picture is obtained then if the Euclidean geometry is used block (13, 14), which replaces the role of modeling, because it gives real micro-structure shapes pictures, on one hand, and from the obtained passive micro-structure picture, through the shape reconstruction, leads to the possibility of its prognoses with designed micro-structure properties block (15). Applying known syntheses phases in the technology process, the obtained samples are investigation objects in these innovations. Thus, the new possibilities in actual solutions and micro-structure characterization application, are directly introduced, whereby the hardware is reduced only to this engineering system component, providing new solutions related to contemporary software support in these devices. Such engineering system in micro-structure fractal application opens industrial production and application possibilities, and their solutions become a base for the *Fractal Electronic* development in the next phases block (16).

### ***Microstructure fractal dimension and analysis***

We strongly stress that the numerical result we may get using eq. (1) is a dimension of the picture we got from SEM, and not of the sample itself. So, it may differ from the real fractal dimension for the same amount the picture differs from the original. It must be taken into account, that automatic counting boxes may also introduce some *numerical noise* as well as the last square approximation, its inherent error. But, in spite of this, results are quite usable in comparing two different samples and are not an obstacle in having insight in complicated processes of sintering and electronic properties of consolidated materials.

Looking back to our earlier investigations [7-11] we may observe several BaTiO<sub>3</sub> ceramics fractality *sources*, as it is anticipated by the points i. to v. in the section *Evidences of fractality*"; namely, the grain contours has  $DH_f$  dimension that lies in the interval 1.1-1.3, calculated by Richardson *variable yardstick* method. The grains' surfaces fractal dimension is estimated to be 2.01 to 2.02. More or less close contacts between individual grains have their own *maps* of touching points having fractal pattern, similar to fingerprint pattern. Similarly, fractal dimension of these touching points lies in the interval 1.6-1.9.

Also, it has to keep in mind that all these data are applied to ceramics material after sintering process is over. It will be of precious interest to follow changing of fractal dimension from *green body's* powder to final ceramics.

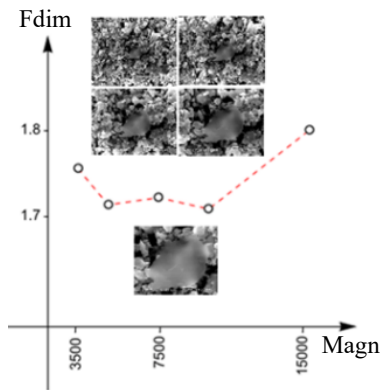
It is worth mentioning that no investigations have been made in estimating fractality of the inside of ceramics bulk. So, with the limitations we spoke, there is what we know now about different issues of fractal dimensions of the ceramic's materials after sintering process.

Analysis of the specimens of BaTiO<sub>3</sub> ceramics doped with Ho are expressed on fig. 3.

## **Results and discussion**

### ***Temperature involvement***

By micro-structures in origin and fractal nature analysis including fractal dimension characterization, any chaotically and disordered structure could be reconstructed. We demonstrate this practical explanation that all chaotically morphologies could be recognized as order or just controlled disorder by fractal self-symmetry approach. Here we experimentally meet



**Figure 3. The diagram of fractal dimension vs. magnification shows variations**

generates a motion in Fermi gas form, containing different particles such as electrons, atoms, atomic nuclei, *etc.* In essence this motion has Brownian character and impose necessity of introducing the third fractality factor – motion factor,  $\alpha_M$  ( $0 < \alpha_M < 1$ ). Our hypothesis is that BaTiO<sub>3</sub> ceramics working temperature must be influenced by these three fractality factors, making correction of *theoretic* temperature as:

$$T_f = \alpha T \quad (5)$$

where  $\alpha$  is fractal corrective factor. It is natural that all three factors  $\alpha_s$ ,  $\alpha_p$ , and  $\alpha_M$ , influences  $\alpha$ :

$$\alpha = \Phi(\alpha_s, \alpha_p, \alpha_M) \quad (6)$$

The argument for this expectation hides in the fact that geometrically irregular motion of huge particles number has to unleash an extra energy to the system. In other words, fractality of system represented by three factors  $\alpha_s$ ,  $\alpha_p$ , and  $\alpha_M$  should increase overall energy of the system, and this increment must be subtracted from the input energy which is in fact, an input thermal energy denoted by  $T$ . In other words,  $T_f = T - VT$  and since it follows from eq. (5) that  $\alpha = T_f/T = (T - VT)/T$ , it yields  $0 < \alpha = 1 - VT/T < 1$ .

More generally, every expression that contains a function of temperature, say  $F(T)$ , if it is differentiable, may be expand to Taylor series:

$$F(T_f) = F(T - \Delta T) = \sum_{k=0}^{+\infty} \frac{1}{k!} F^{(k)}(T) (-1)^k \Delta T^k \quad (7)$$

The nature of the function  $\Phi$  in eq. (6) is unknown by now, but in the first moment, the linear approximation will suffice:

$$\Phi(\alpha_s, \alpha_p, \alpha_M) = u\alpha_s + v\alpha_p + w\alpha_M \quad (8)$$

where  $u, v, w > 0$  are real coefficients satisfying  $u + v + w = 1$ .

Although fractal dimension of the ceramics volume is still under the research, we might be quite sure that it is between 2.5 and 3. In the manuscripts [15] we introduce the formulas for calculating fractal dimension  $DH_f(t)$  as a function of sintering time,  $t$ , in all three sintering phases.

The practical value is that the fractal objects interaction and energy is possible at any reasonable scale of magnitude, including nanoscale as a consequence of fractal independence on

possibility that disorder is transformed by self-similarity to structures in order. So, that is the *hotline* where fractals meeting entropy as measure of the chaos in the structures. We will here introduce the temperature as important thermodynamic parameter which influences on entropy and materials structure morphology. Arguing about the crystal surface *natural roughness* as macroscopic steps collection on the arbitrary section surface of the crystal plane section, the authors of [12-14] quote an observation Frenkel had come forward with, that this roughness does not coincide with the crystal faces atomic roughness, with small surface energy, which can occur as a thermal fluctuations consequence at high temperatures. This temperature consideration illustrates impact on dynamical processes inside the ceramics body, which

scales. The relation energy-structure-fractals in correlation with entropy is the question of central importance for this research and it is scientifically very challenging and biunivocally involving at the same time. If we consider two contra positioned energy questions, surface tension, which measures the energy needed to create a surface, (for example makes the soap film surface in free space to take spherical form) which deforms film to minimize its surface and, thus its energy in relation with order to supply live tissues uniformly, directly lead to existing various types of chaotically structures (drain capillaries, veins and vessels, *etc.*). So, the energy, energy stability, structures and orders-disorders are naturally connecting the fractal nature and entropy phenomena's.

**Entropy and fractals in nature, entropy, and fractal dimensions**

Between the thermodynamic fundamental parameters, Gibbs free energy, enthalpy, entropy, the entropy-temperature relation is of enormous importance.

Because the main subject of our paper is entropy-temperature-structures-fractals relation we will continue to analyse this priority goal.

*Threshold identification using maximum entropy*

For each relevant class identified to define a valley, a threshold is calculated based on entropy, considering a probability of an intensity correctly segmenting a given group of objects. For such, the image is taken as the result of a random process in which probability  $p$  corresponds to the probability of a pixel in the image taking on an intensity value  $i$  ( $i = 1, \dots, n$ ), as shown in eqs. (1) and (2). The intensity or grey level of the class with the greatest entropy is identified as a threshold:

$$S = - \sum_{i=1}^n p_i \log(p_i), \quad p_i = \frac{n_i}{N} \tag{9}$$








in which  $S$  is the Shannon entropy of the image,  $n$  – the total number of outputs (number of grey levels in the image),  $p_i$  – the probability of grey level  $i$  being found in the image,  $n_i$  – the number of pixels with intensity  $I$ , and  $N$  – the total number of pixels in the image.

As an illustration, the central grain of doped BaTiO<sub>3</sub> ceramics shown at the SEM photo below, fig. 4, will be used to evaluate its Shannon entropy upon different resolutions.

*Information dimension DH<sub>i</sub>*

The simplest entropy-based fractal dimension is related to the first-order Shannon entropy. Let us consider an arbitrary fractal that is covered by  $N$  veils, each with a diameter  $r$ , at the  $k^{\text{th}}$  covering (which is a setting similar to that used to determine the Hausdorff dimension,  $DH$ ). Recall that  $DH$  was estimated from the number of veils intersected by the fractal, regardless of the density of the fractal in each veil. In contrast, the estimation of the information dimension,  $DH_1$  considers the density of the fractal, as determined from the relative frequency of occurrence of the fractal in each intersecting veil. If  $n_{jk}$  is the frequency with which the fractal enters (intersects) the  $j^{\text{th}}$  veil of size  $r_k$  in the  $k^{\text{th}}$  covering, then its ratio to the total number  $NT_k$  of intersects of the fractal with all the veils is an estimate of the probability  $p_{jk}$  of the fractal within the  $j^{\text{th}}$  veil, and is given by:

$$p_{jk} \stackrel{\text{def}}{=} \lim_{k \rightarrow \infty} \left( \frac{n_{jk}}{N_{Tk}} \right), \quad N_{Tk} \stackrel{\text{def}}{=} \sum_{j=1}^{Nk} n_{jk} \tag{10}$$

						
$N=30976$ $i \leq 22655$ $S=9.83071$	$N=7744$ $i \leq 5877$ $S=8.48521$	$N=1935$ $i \leq 1371$ $S=7.04221$	$N=484$ $i \leq 396$ $S=5.77977$	$N=121$ $i \leq 102$ $S=4.4992$	$N=36$ $i \leq 28$ $S=3.26092$	$N=9$ $i \leq 5$ $S=1.52296$

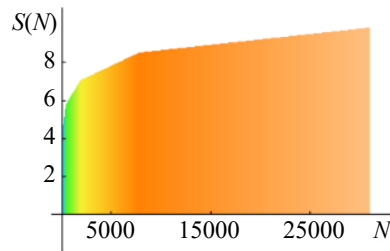


Figure 4. Shannon entropies from the table

Notice that this total number  $N_{T_k}$  must be recalculated for each  $k^{\text{th}}$  covering because, in general, it can change substantially on dense fractals. With this probability distribution at the  $k^{\text{th}}$  covering, the average (expected) self-information *i. e.*,  $I_{jk} = \log(1/p_{jk})$  of the fractal contained in the  $N_k$  veils can be expressed by the Shannon entropy  $S_{1k}$  as given:

$$S_{1k} \stackrel{\text{def}}{=} - \sum_{j=1}^{N_k} p_{jk} I_{jk} = - \sum_{i=1}^{N_k} p_{jk} \log(p_{jk}) \tag{11}$$

Notice that the subscript 1 in  $S$  denotes that the Shannon entropy is of the first order which assumes independence between all the veils. If the following power-law relationship holds:

$$c^{S_{1k}} = K \left( \frac{1}{r_k} \right)^{DH_I} \tag{12}$$

where  $c$  and  $K$  are the constants, then the information fractal dimension is:

$$DH_I = \lim_{k \rightarrow \infty} \frac{S_{1k}}{\log \left( \frac{1}{r_k} \right)} \tag{13}$$

*Correlation dimension  $DH_c$*

The information dimension is an extension of the Shannon-entropy-based dimension. It reveals the expected spread in the non-uniform probability distribution of the fractal, but not its correlation. The correlation fractal dimension was introduced to address this problem. Let us consider a setting identical to that required to define the information dimension,  $DH_I$ . If we assume the following power-law relationship:

$$\left( \sum_{j=1}^{N_k} p_{jk}^2 \right)^{-1} = K \left( \frac{1}{r} \right)^{DH_C}, \quad K \text{ is constant} \tag{14}$$



where from yields:

$$DH_C = \lim_{k \rightarrow \infty} \frac{-\log \sum_{j=1}^{N_k} p_{jk}^2}{\log \left( \frac{1}{r_k} \right)} \quad (15)$$

*Renyi dimension spectrum  $DH_q$*

Introduced by Renyi in 1955 [16], the generalized entropy is given:

$$S_q = \frac{1}{1-q} \log \left( \sum_{j=1}^{N_k} p_{jk}^q \right), \quad 0 \leq q \leq \infty \quad (16)$$

Parameter  $q$  is called moment order.

Since continuously depends on moment order  $q$ , it is used called Renyi spectrum, and the graph of this dependence is given the next picture, fig. 5.

It shows monofractals, *i. e.* the fractals having constant fractal dimension all over the fractal surface. On the contrary, multifractals vary  $DH$  from point to point, which may serve as definition of a multifractal.

If the values of moment order extend to negative real axis, so  $-\infty \leq q \leq +\infty$  than the power law takes place:

$$\left( \sum_{j=1}^{N_k} p_{jk}^q \right)^{\frac{1}{1-q}} = K \left( \frac{1}{r} \right)^{DH_q} \quad (17)$$

where from the Renyi dimension follows:

$$DH_q = \frac{1}{1-q} \lim_{k \rightarrow \infty} \frac{\log \sum_{j=1}^{N_k} p_{jk}^q}{\log \left( \frac{1}{r_k} \right)} = \frac{1}{q-1} \lim_{k \rightarrow \infty} \frac{\log \sum_{j=1}^{N_k} p_{jk}^q}{\log(r_k)} \quad (18)$$

$$DH_q = - \lim_{k \rightarrow \infty} \frac{S_q}{\log(r_k)} \quad (19)$$

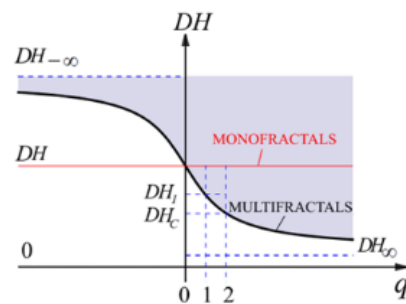


Figure 5. Renyi Spectrum for multifractals

**Conclusions**

Analysis of the contour of the real BaTiO<sub>3</sub> ceramics grain is an important example which shows a few features of fractal approach: gives much more natural approximation to modern technology real physical structures than classic Euclidean geometry, fractals are not just geometric objects *e. g.* processes parameters may have fractal behaviour in time (changing of energy, motion of particles (Brownian walk), varying of electric or mechanic parameters, *etc.*), fractal dimension offers a mighty instrument for fine distinguish and precise identification of processes that may look identical at the first sight.



**Figure 6. The interrelations overview: Space-Earth-Nature-Energy-Fractals-Entropy**

We establish relation between the fractals and entropy and to open new frontiers in order to define what the order is and how to control it. Our aim could be the idea, that disordered and chaos of many structures, especially micro-structures, could be controlled and renewed. The relation between entropy and fractals on the Earth and in total in Universum is also very important for the basic energy questions, fig. 6. Developed idea bridgeing entropy and fractals is fundamental for future frontiers in this area and by fractals thermodynamic science is getting more advancement within the modern science.

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