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Fractal modification of Curie-Weiss law based improved impedance of doped $BaTiO_3$-ceramics

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Abstract

A new approach based on fractal geometry and correlation between microstructure-nanostructure and rare-earth properties and other additives doped $BaTiO_3$-ceramics and electronics properties, is applied. The grain contacts geometry based on different stereological models and especially intergranular contact surface fractal morphology was the subject of several papers of these authors [7-34]. The main conclusion was that intergranular capacities have higher values then expected which is induced by contact surfaces sizes augmentation as a consequence of their fractal nature. Bigger micro-capacities was justified by virtually having bigger dielectric constants which was performed by introducing *fractal correction factor* $a_0 > 1$, as a multiplier to the usual dielectric constant e_r to gain the bigger value $a_0 e_r$. In further $BaTiO_3$-ceramics micro-electronics fractal theory development, different fractal methods were used to describe complexity of the spatial distribution of $BaTiO_3$-grains, as well as pores [13-15]. This led to fractal correction factor revisited model, which considers existence of surfaces fractality, pores fractality and the inner liquid dynamics and solid state sintering phase within ceramics material represented by the Brownian motion model. The new correction factor a is a function of these three ceramics fractality "sources". Here, the relationship between a and a_0 is established for doped $BaTiO_3$-ceramics using the Curie-Weiss law and temperature as an omnipresent sintering parameter. Also, the model of two grains contact impedance is studied for different a and frequency values. By the control of shapes and contact surfaces numbers on the the entire $BaTiO_3$-ceramic sample level, the control over structural properties of these ceramics can be done, with the aim of correlation between material electronic properties and corresponding microstructure. The fractal nature for analysis of the ceramics structure providing a new approach for modeling and prognosis the grain shape and relations between the $BaTiO_3$-ceramic structure, micro- and nano-, and all other electronic properties in the light of new frontier for higher level electronic circuits integration.

Keywords: **$BaTiO_3$-ceramics, fractal, microstructure, micro-capacity, micro- impedance**.

Introduction

Barium-titanate ceramics are one of the most important electronic ceramics for the small size and multilayer capacitors of high capacitance manufacture. For these applications this ferroelectric is usually doped with various additives such as Er^{+}, Ho^{3+}, Mn^{2+}, Nb^{5+}, Yb^{3+}, and some oxides, in attempt to achieve a temperature stable dielectrics. It is shown that dielectric constant depends strongly on grain and pore size in the ferroelectric state, i.e. the finer the grain size, the higher the dielectric constant. The investigation of microstructure characteristics of undoped and doped $BaTiO_3$ in function of consolidation parameters is a necessary step in barium-titanate ceramics processing and designing. Structure investigations provide better understanding for dialectic properties, especially from the point of view of the relative dielectric constant response of pure and doped $BaTiO_3$-ceramics. Fractal method traces a new approach for describing and modeling the grain's shape and relations between $BaTiO_3$-ceramics structure and electrical properties. It gives more natural approximation to the grain's boundary, but the construction uses recursive random algorithms. Particle shape is a fundamental powder property, affecting powder packing and thus bulk density, porosity, permeability, cohesion, flowability, attrition, interaction with fluids, covering power of pigments, resistance, capacity, magnetic permeability etc. Introducing mathematically well-established fractal sets theory in the powder metallurgy science, new materials, nano-technology, “free-floating” metallic drops processing, various ceramics technologies, graphene oxide flakes, literally from trash recycling to self-healing materials.

Using the fractal theory modern developments, offers enough firm arguments to support modelling, predicting and many modern technological processes control, as outstanding examples of fractal-based structures. Estimation the fractal analysis main parameter - the Hausdorff or fractal dimension, for all relevant morphologies that appear in sintering processes. By using obtained values, study their impacts on distribution of energy, temperature, surface tension, dielectric constants, rate of densification etc. The fractal analysis is used in ceramics materials to quantify the particle's structure complexity, granular complexes, sintering processes, pores distribution changes etc.

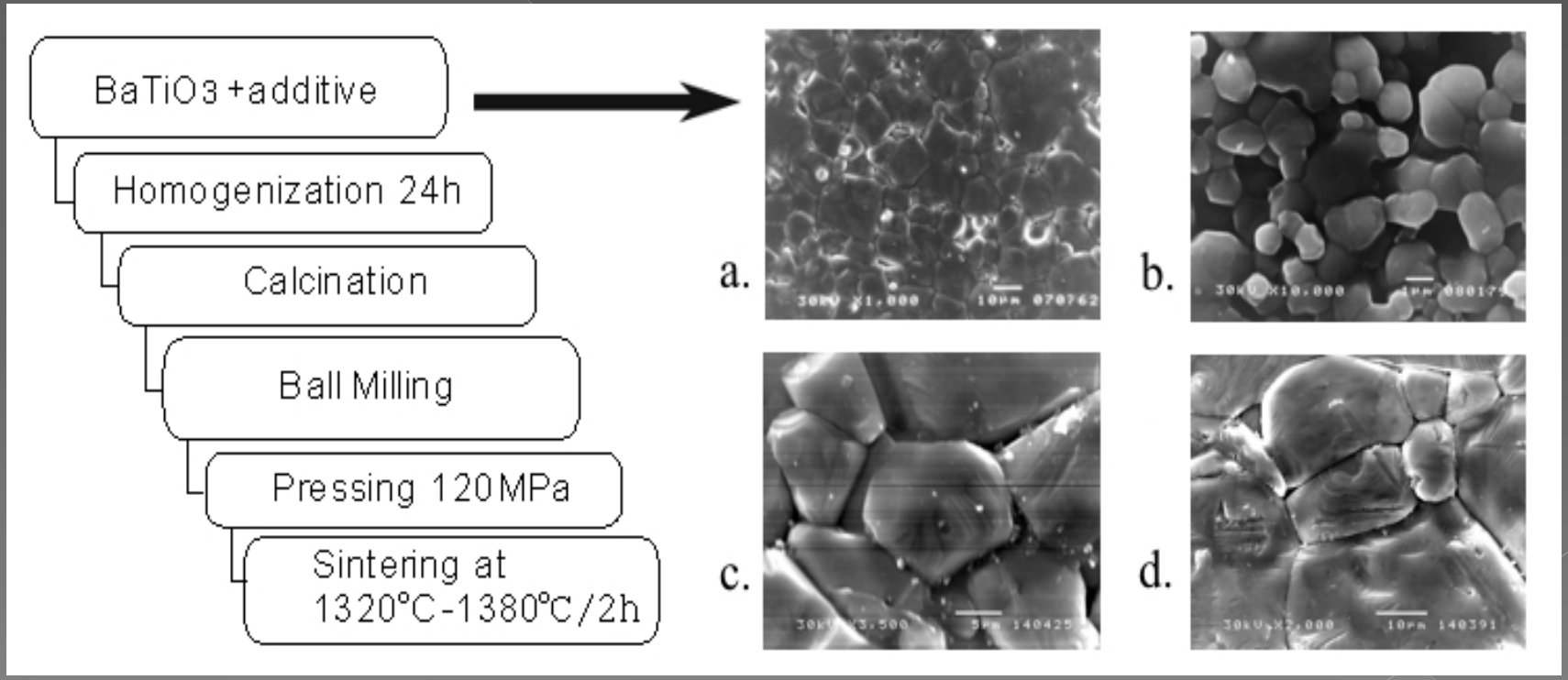


Fig.1. Doped BaTiO3 powder preparation. **a.** 0.5wt%Er/1320 ^{o}C; **b.** 1.0wt%Yb_2O_3/1350 ^{o}C; **c.** 0.01wt% Ho/1380 ^{o}C; **d.** 0.1wt%Ho/1350^{o}C;

Experimental procedure

In this paper, Er_2O_3, Ho_2O_3, Mn_2O_3, Nb_2O_3, Yb_2O_3, doped $BaTiO_3$-ceramics were used for microstructure characterization, and modeling.

The samples were prepared from high purity (>99.98%) commercial $BaTiO_3$ powder (MURATA) with [Ba]/[Ti]=1,005 and Ho_2O_3 powders (Fluka chemika) by conventional solid state sintering procedure. The content of Ho_2O_3 ranged from 0.50 to 2.0 wt% .

Starting powders were ball-milled in ethyl alcohol for 24 hours. After drying at 200ºC for several hours, the powders were pressed into disk of 7mm in diameter and 3mm in thickness under 120 MPa.

The compacts were sintered from 1320ºC to 1380ºC in air for four hours. The microstructures of sintered and chemically etched samples were observed by scanning electron microscope (JEOL-JSM 5300) equipped with energy dispersive spectrometer (EDS-QX 2000S).

Fractality and ceramics grains' contacts

Electronics ceramics, especially BaTiO3-ceramics, are made out of very fine powder having the maximum Ferret diameter. These particles have so high surface energy to fuse together and to make sintered ceramics. As it is well known[1-6], many powder materials have fractal structure, and nowadays it is well established, documented and widely accepted fact (Figure 2). Fractal geometry, systematically introduced by Benoit Mandelbrot[1,2] during the end of sixth and at the beginning of seven decade of the last century (for a constructive approach, see Barnsley[3]) as an efficient toll for describing complex non-Euclidean shapes. *Shapes which are not fractal are the exception*, said Mandelbrot. The fractal geometry key concept is the unique number that is connected with fractal object which is known as *fractal dimension*, a concept introduced sixty years before by Felix Hausdorff. If DH_f denotes fractal dimension, the simple inequality $DH_f > DT$, where DT is topological dimension, has been suggested by Mandelbrot as an acceptable (although not complete) definition of fractal objects.

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2. B. Mandelbrot, Les objets fractals, forme, hasard et dimension, Flammarion, Paris, 1975.
3. M. Barnsley, Fractals Everywhere. Academic Press, 1988.
4. B. H. Kaye, A Random Walk Through Fractal Dimensions, Wiley-VCH 1994.
5. A. Naman, Mechanical Property and Fractal Dimension Determination in Li2O*2SiO2 Glass-ceramics, University of Florida, 1994.
6. N. M. Brown, The Fractal Dances of Nature, Penn State News, Wednesday, November 19, 2014.

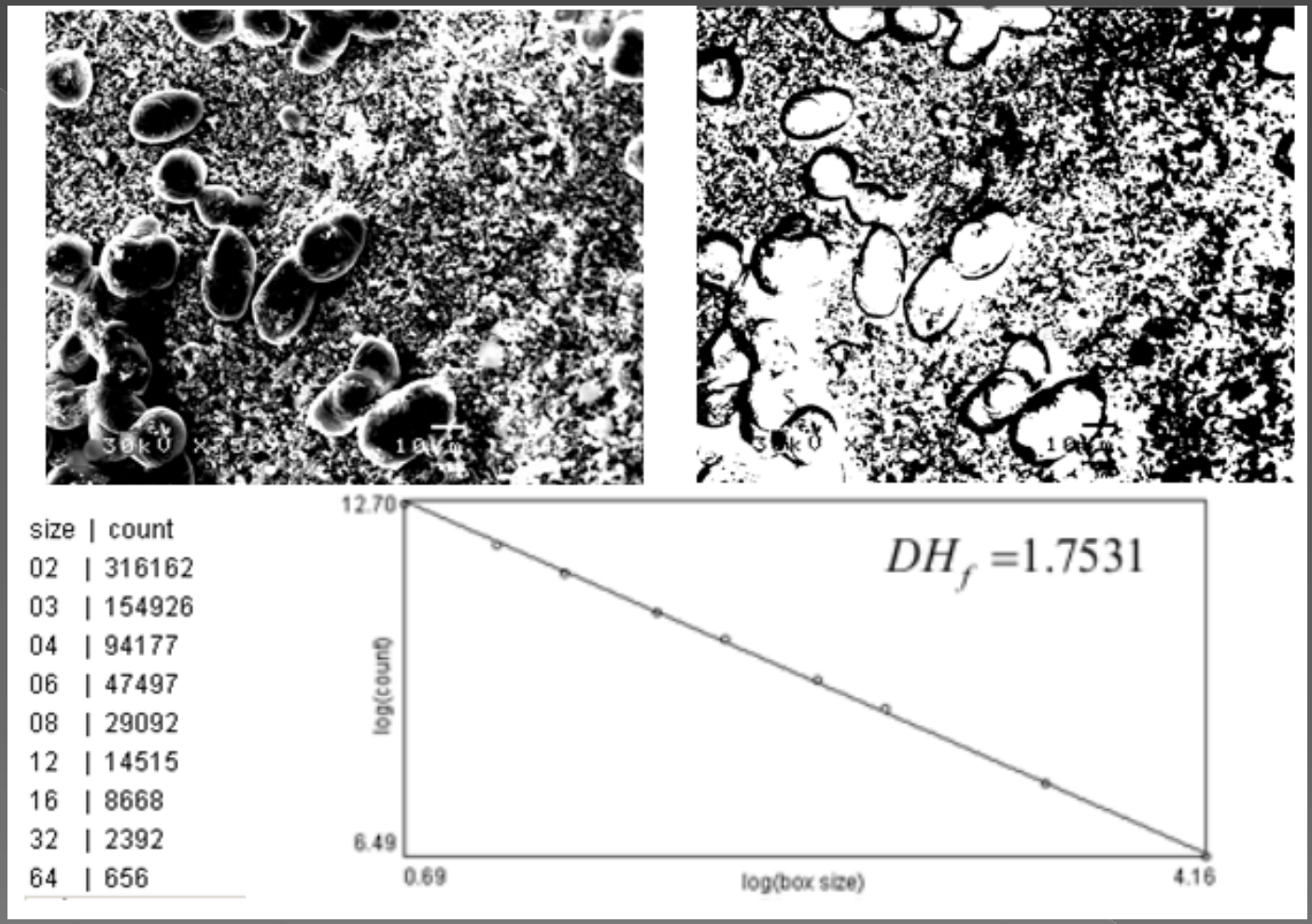


Fig. 2. *Above*. The $BaTiO_3$ doped with 0.5wt%Ho ceramics, sintered on 1320 ºC/120 MPa for 4 hours SEM sample (left) and its BW electronic image; *Below*: Fractal dimension of the sample extracted by gray level box counting method gives $DH_f = 1.7531$.

Jack Mecholsky, a professor at Penn State expressed his attitude towards a new science: „Fractal geometry is probably the most useful math ever invented.” In 1976, Mecholsky, then at Sandia National Labs, together with Dann Passoja were among the first trying to apply new fractal geometry on metal fracture surfaces. In 1984 Mecholsky suggested a fractals project for her senior thesis in ceramic science. He, Passoja, and Karen Feinberg took samples of brittle ceramics, broke them, and measured the fractal dimension of the fracture surface [*Brown*]. They found that the toughness of the material was proportional to the fractal dimension,

$$K_{Ic} \sim \frac{1}{2}\left\langle DH_f \right\rangle$$

where K_{Ic} is toughness and $\left\langle DH_f \right\rangle$ is the decimal part of the fractal dimension.

Applied to $BaTiO_3$ and similar perovskite ceramics, a grain contour line typically satisfies $1.06 < DH_f$ (contour) < 1.08 [7,8] which is much in agreement with other author's measurements results (see Smirnov [9] , p.38). The DH_f value depends on the sintering phase, and for the surface of a grain it is $2.079 < DH_f$ (grain's surface) < 2.095 [10] . Many considerations and numerical work has been spent in two-grain contact area size approximate evaluation. Different models were studied having supposition that one or both grains belong to one of three approximate classes: polyhedrons or prisms (P), spheres (S) or ellipsoids (including spheroids) (E). All possible two grains contact combinations have been considered: P-P, S-S, E-E, P-S, P-E and S-E [10-12] .

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7. V. V. Mitić, Lj. M. Kocić, M. M. Ristić, The Interrelations between Fractal and Electrical Properties of BaTiO3-Ceramics, The American Ceramic Society 99th Annual Meeting and Exposition, Cincinnati, Ohio, May 4-7, p. 199, 1997.

8. Mitić V.V., Kocić M.Lj., Miljković M., Petković I., Fractals and BaTiO3-Ceramic Microstructure Analysis. Mikrochim. Acta 15, 365-369, Springer-Verlag (1998).

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11. P. Petković, V. V. Mitić, Lj. Kocić, Contribution to BaTiO3-Ceramics Structure Analysis by Using Fractals, Folia Anatomica, 26, Suppl. 1, pp. 67-69, 1998.

12. V. V. Mitić, Lj. M. Kocić, I. Z. Mitrović, M.M. Ristić, Shapes and Grains Structures Stochastic Modelling in Ceramics, Proc. of S4G - International Conference on Stereology, Spatial Statistics and Stochastic Geometry, Prague, Czech Republic, June 21-24, pp. 209-215, 1999.

Using the Mandelbrot's formula that gives a relationship between the length L of a fractal "space filling" curve of fractal dimension DH_f that "fills" the two-dimensional area of size A , i.e.,

$$L^{-DH_f} = K\sqrt{A}, \quad (K \text{ is constant}),$$

in combination with Richardson law of variable yardstick, upon which a fractal curve length depends on measurement precision, i.e. on the measure yardstick length δ is

$$L = \delta^{1-DH_f},$$

gives A as a function of DH_f

$$A(DH_f) = K\delta^{2\left(\frac{1}{DH_f}-1\right)}. \qquad (1)$$

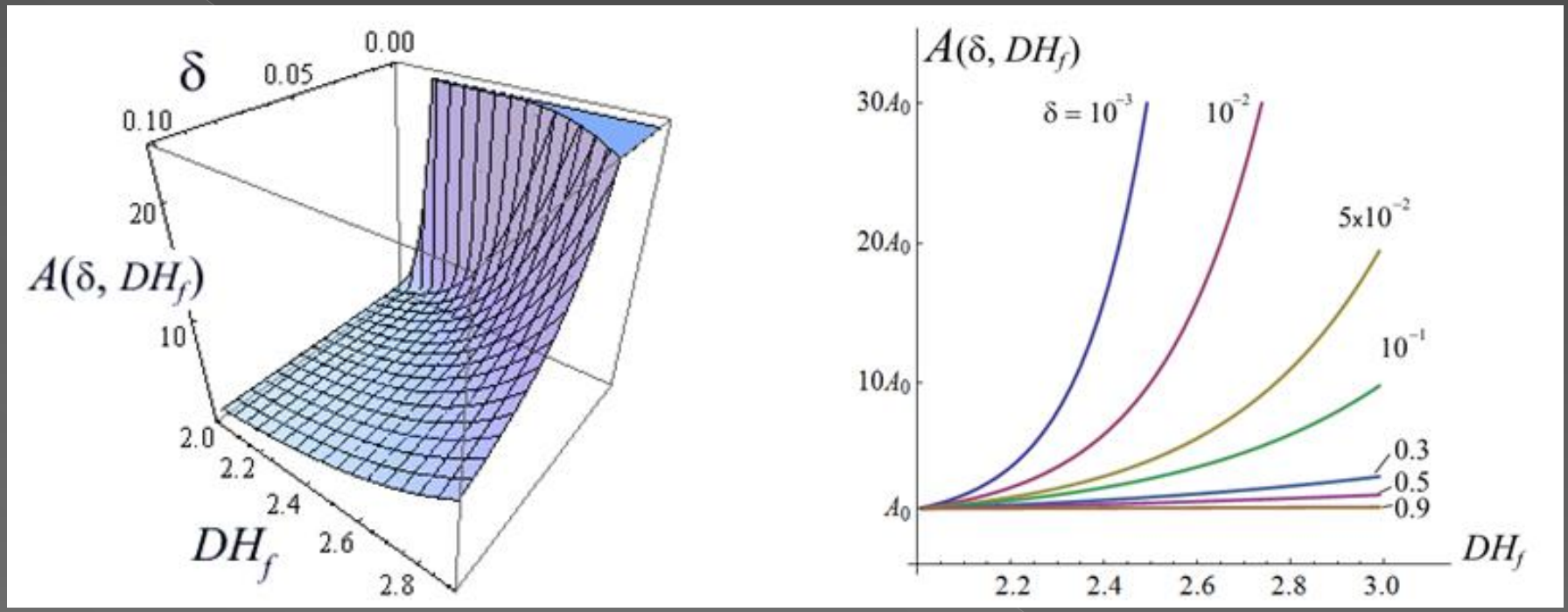


Fig. 3. Size of the contact area $A(\delta, DH_f)$ vs. fractal dimension. A_0 in the right 2D graph is the area size for ideally flat contact surface.

Such considerations did help in gaining the approximate value of neck cross section area size *A* in Coble two grain contact model, where real cross section is approximated by *a*, *b* semi-axes elliptic disc as

$$A_{\text{Coble}} = 2ab\pi\,\delta^{2\left(\frac{1}{DH_f}-1\right)}.$$

From the last two formulas is clear that the surface area increases when □ gets smaller; theoretically, for $1 < DH_f$, $A \to +\infty$ when $\delta \to 0_+$.

In addition, the area of ceramics surface (as a cluster of grains) as well as the area of a single grain surface, depends on the unit of measure δ and local fractal dimension DHf and is given by

$$A(\delta, DH_f) \sim \delta^{2-DH_f}. \qquad (2)$$

The relationship (2) is illustrated by the graphs at Figure 3. The increasing of the contact area size with more precise measuring (smaller *d*) is evident for all fractal dimensions > 2. Even for „smooth“ fractal surfaces, i.e. surfaces with DH_f close to 2 (as it is the case with $BaTiO_3$-ceramics grains surfaces having $2.079 < DH_f < 2.095$), the area size *A* duplicates its value if the unit of measure *d* decreases for the factor 2.87389×10^{-4} .

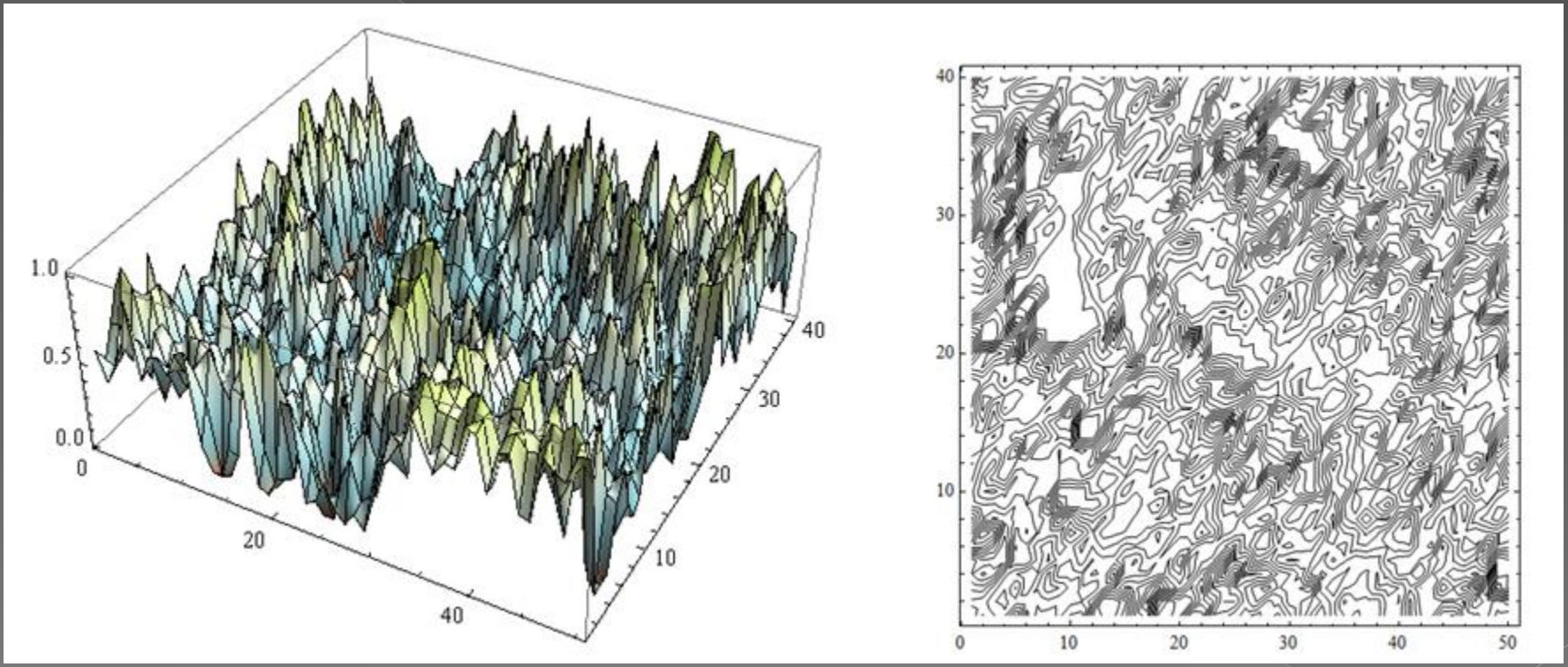


Fig. 4. Left. Reconstruction of 3D-surface for $BaTiO_3$ sample from Fig. 2; Right. Level lines of the same surface. Picks represent grains.

While the fractal dimension of $BaTiO_3$-ceramics grain's surface is modest, the dimension of the specimen surface is much higher, as Figure 4 shows. The fractal dimension is calculated using max-gray level box-counting method of the SEM, which yields $DH_f = 1.7531$. For other specimens similar values are obtained in range from 1.7529 to 1.8025.

These results led to revision of the formula for parallel-plate capacitor with plates area A and a separation d ($d << A^{1/2}$)

$$C = \varepsilon_r \varepsilon_0 \frac{A}{d} \tag{3}$$

where e_r and e_0 are dielectric constants of $BaTiO_3$-ceramics grains' contact zone and in vacuum respectively. Namely, the intergranular microcapacitor, formed in the contact zone of two ceramics grains is *not* a parallel-plate capacitor. It is a fractal capacitor that may be thought of as being product of a iterative process described by the Figure 5, left. The top subfigure shows a parallel-plate capacitor as described above. It corresponds to a capacitor (denoted by C_0) with adjacent grains' perfectly flat contact surfaces which does not exist in reality. On the contrary, the contact surface is rough and uneven, so that the following fractal model will be a good approximation. Suppose that the flat parallel geometry of C_0 (Figure 5, left) is replaced by three flat capacitors „Z"-shaped configuration connected in parallel, forming the unique capacitor C_1.

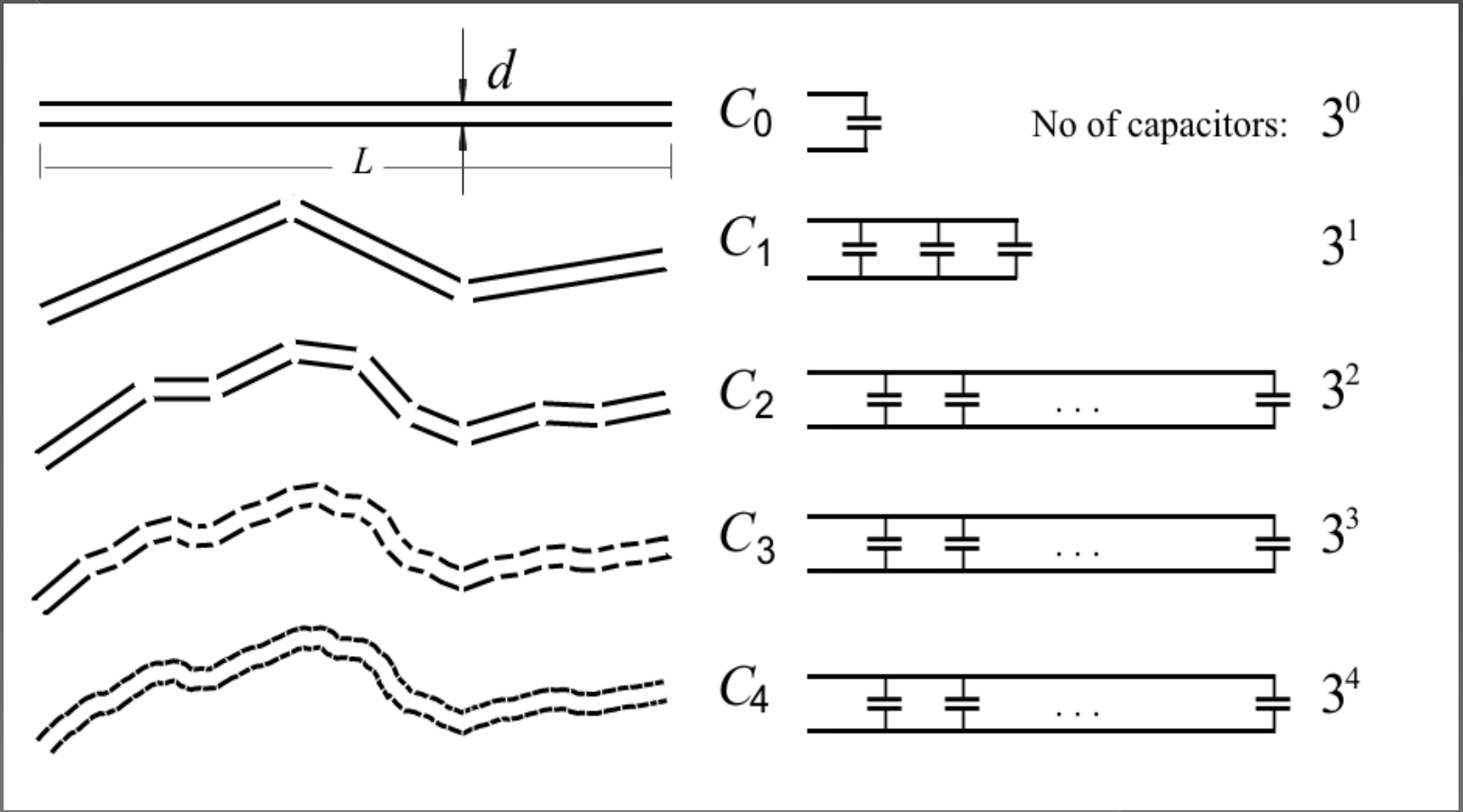


Fig. 5. Constructive way to explain fractal character of an intergranular microcapacitor.

In the next stage, each of three linear segments is replaced by another smaller „Z“ made out of three smaller flat capacitors. By continuing this procedure, the sequence of more and more segmented chains of sub-capacitors C_0, C_1 , C_2,..., is obtained (Figure 5, right). It is suitable to take each smaller „Z“ to be a shrunk *affine image* of the bigger „Z“ standing in the above line. In this manner, the separating distance would be shrunk as well for some factor (although it is not shown in our figure for clarity reasons). This factor (called vertical scaling factor), makes the ratio $d/\sqrt{A}$ smaller for each iteration, making capacity to increase. Also, the sum of the lengths in each iteration increases due to wiggling form of the higher stage capacitor. In all, the capacity of C_n will be substantially bigger than C_0. This increment is characterized by the ratio $\alpha_0 = \lim_{n\to\infty} C_n / C_0$, and thou, if C_f stands for “fractal capacitor”

$$C_f = \lim_{n\to\infty} C_n = \alpha_0 \varepsilon_r \varepsilon_0 \frac{A}{d} = \alpha_0 \left(\varepsilon_r \varepsilon_0 \frac{A}{d} \right) = \alpha_0 C \qquad (4)$$

where C is capacity of the capacitor having ideally flat plates.

Since, $C_f > C$ it follows that $a_0 > 1$. This effect of increasing capacity due to fractality of contact zone is referred by these authors as the *a-correction* of the intergranular capacity or, which is the same, dielectric constant, by stating [13, 14]

$$\varepsilon_{r\,f} = \alpha_0 \varepsilon_r$$

Indeed, the increasing in capacity is the consequence of micro-structure, not of macro-parameters like grains' position or size. Consequently, it may be consider as an intrinsic $BaTiO_3$-ceramics characteristic and also doped $BaTiO_3$-ceramics.

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13. V. V. Mitić, Lj. Kocić, Z. I. Mitrović, Fractals and BaTiO3-Ceramics Intergranular Impedance, Gordon Research Conference: Ceramics, Solid State Studies in, Kimball Union Academy, Meriden, New Hampshire, August 1-6, 1999.

14. V. V. Mitić, Lj. Kocić, Z. I. Mitrović, Primena fraktala u modelu intergranularne impedanse BaTiO3-keramičkih materijala. Naučni skup: "Trijada Sinteza-Struktura-Svojstva-Osnova tehnologije novih materijala", Beograd, Novembar 16-18, 1999, str. 65-66.

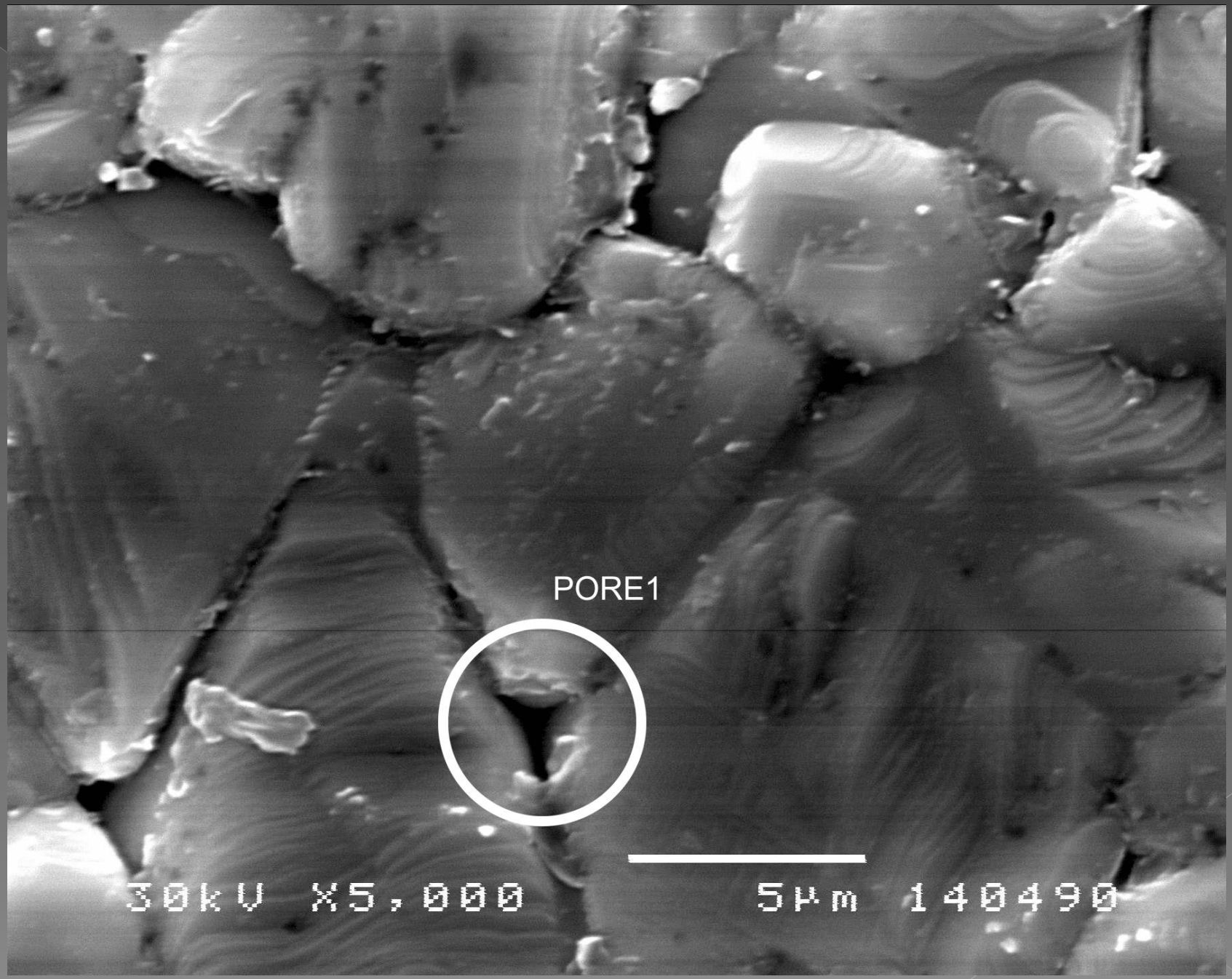
Geometry of pores

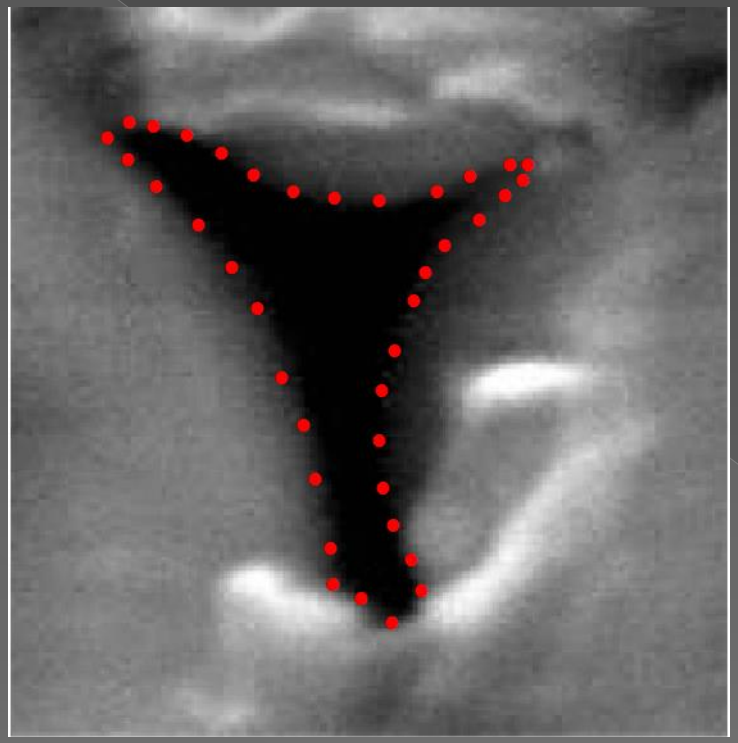
As Gaspard [15] noticed, second law of thermodynamics seems to be in contradiction with the Liouville theorem of Hamiltonian dynamics phase-space volumes preservation. In fact, the Liouville equation describing the probability $\rho_{p,q}(t)$ evolution in time

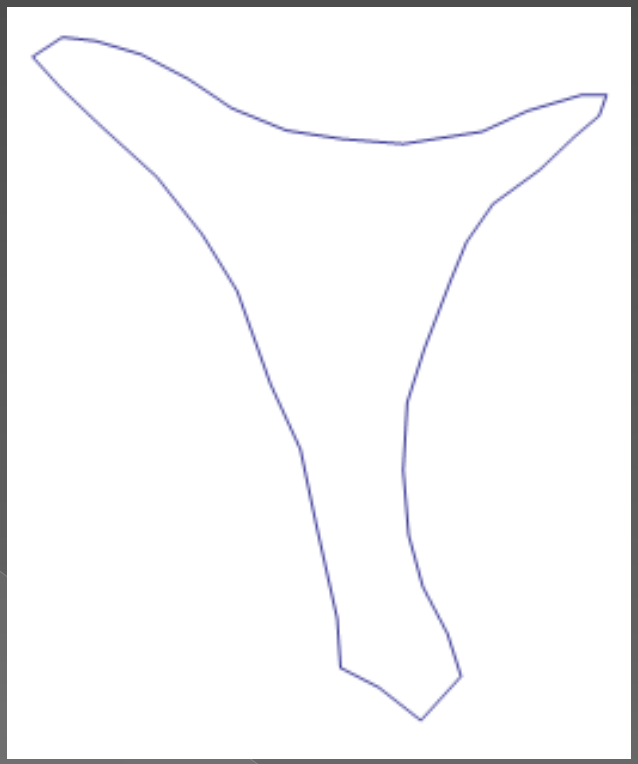
$$\frac{\partial \rho}{\partial t} = \sum_{i=1}^{n} \left(\frac{\partial \rho}{\partial p_i} \dot{p}_i + \frac{\partial \rho}{\partial q_i} \dot{q}_i \right),$$

demonstrates the density conservation in phase space for irreversible processes such as diffusion, the entropy increases in a monotonous manner. Gaspard continues ([15]): "As long as a system is described by regular distributions with a differentiable cumulative function, Liouville's theorem has for consequence that the entropy temporarily remains constant in time. However, if the relaxation dynamics towards the thermodynamic equilibrium involves hydrodynamic modes described by singular distributions with a nondifferentiable cumulative function, we may expect that the entropy no longer remains constant because of the singular and the hydrodynamic modes fractal character."

15. P. Gaspard, Chaos, Fractals And Thermodynamics, Bulletin de la Classe des Sciences de l'Acad´emie Royale de Belgique, 6e s´erie, tome XI (2000) 9-48.

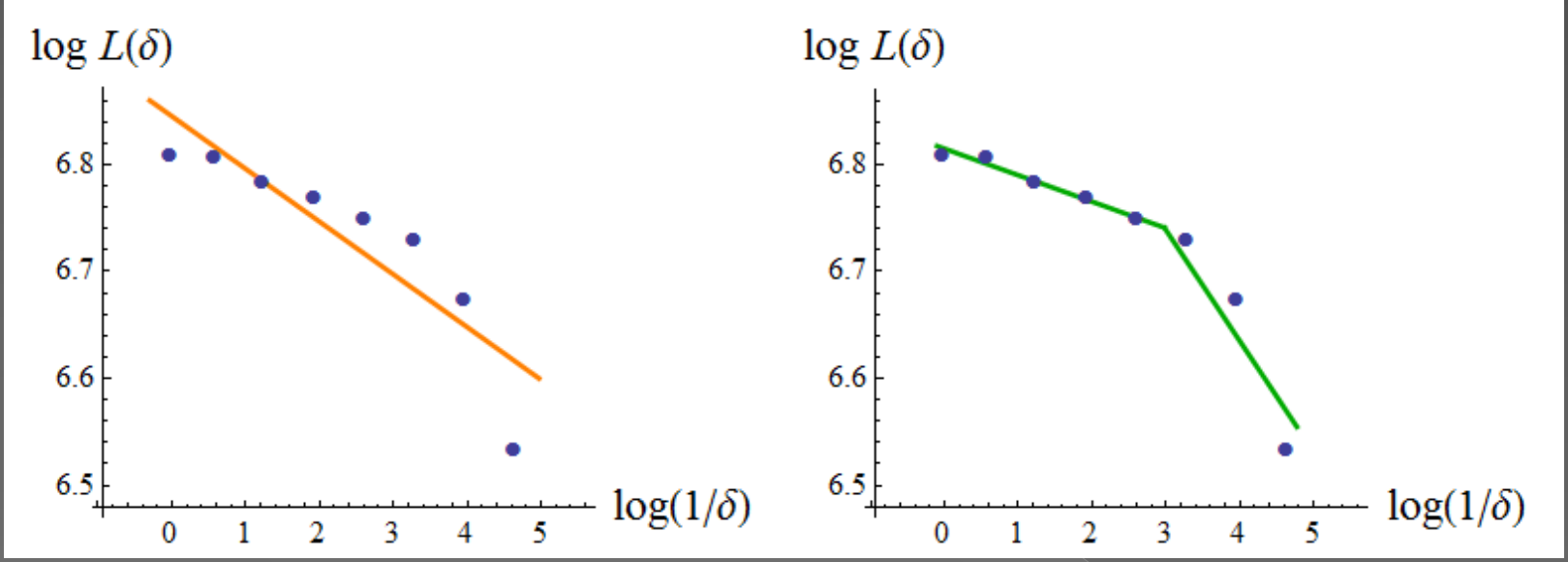

PORE1
30kV X5,000
5µm 140490





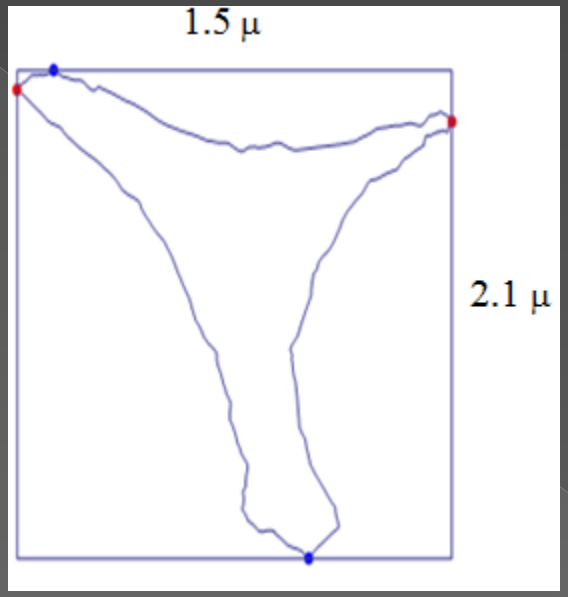
The chosen pore is processed by sampling its contour points to get a polygonal representation that gives the approximately pore's outline.

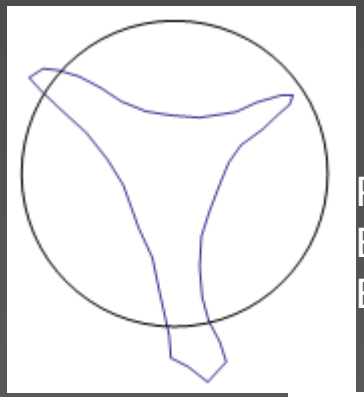
The Richardson method [1] and Kaye's modification [4] , help to retrieve the approximate fractal dimension of the pore's outline.



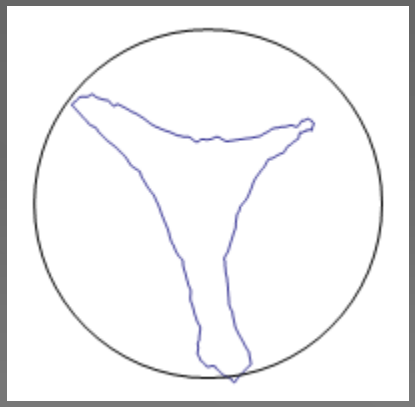
Using 8 different "yardstick" lengths the eight points are obtained in the log-log diagram, gives DH_f = 1.0493 ;

The modification of Brian Kaye gives DH_f = 1.06427 as the average of 1.02478 (upper, "textural" slope and lower, "structural", more slanted slope1.10376.

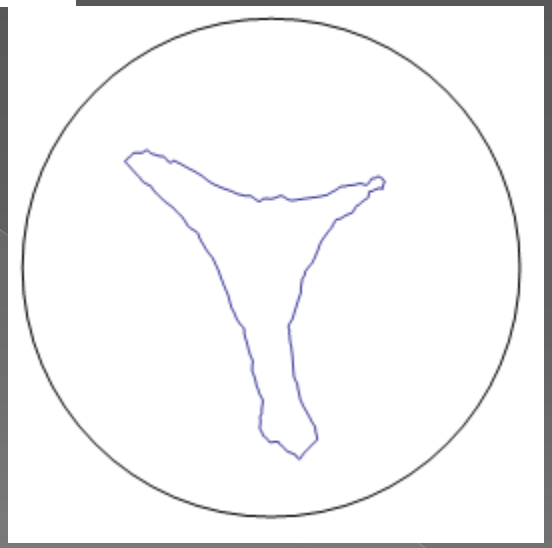




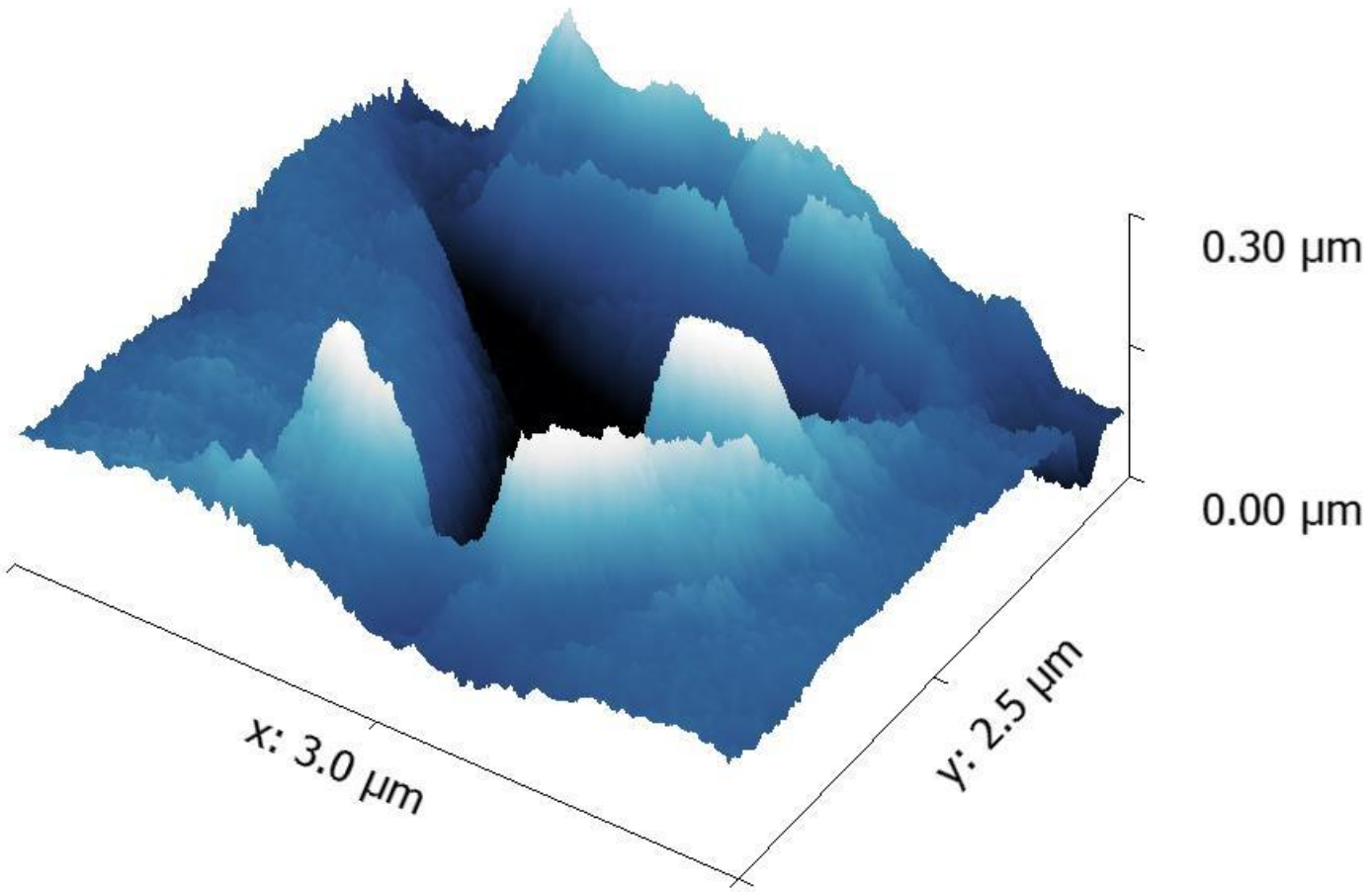
Polygonal approximation:
Equivalent circle radius = 0.8636 μ
Equivalent perimeter = 5. 42624 μ



3rd pre-fractal approximation:
Equivalent circle radius = 1.07818 μ
Equivalent perimeter = 6.77442 μ



5th pre-fractal approximation:
Equivalent circle radius = 1.43756 μ
Equivalent perimeter = 9.03245 μ



micro level. Knowing the fractal dimension (even approximately) helps getting a more realistic model that may explain many micro-processes nature.

Behavior of pores wall fractal dimension durin sintering process:

$$DH_f(t) = \lim_{\varepsilon \to 0} \frac{\ln\left(N_0(\varepsilon) - \frac{3\pi}{4\varepsilon^3}\left(R_0 - \frac{4}{3}\frac{\sigma}{\eta}t \right)^3 \right)}{\ln(1/\varepsilon)}$$

1-st stage of viscous sintering

$$DH_f(t) = \lim_{\varepsilon \to 0} \frac{\ln\left(N_0(\varepsilon) - \frac{4\pi t}{3\varepsilon^3}\left(\frac{\gamma n^{1/3}}{\eta} \right) \frac{\left(\pi - 4\sqrt{2}R_0 \right) R_0^{-2/3} R(t)}{\left(3\pi - 8\sqrt{2}R_0 \right)^{2/3}} \right)}{\ln(1/\varepsilon)}$$

2-nd stage of viscous sintering

$$DH_f(t) = \lim_{\varepsilon \to 0} \frac{\ln\left(N_0(\varepsilon) - \frac{t}{2\varepsilon^3}\left(\frac{4\pi}{3} \right)^{1/3}\left(\frac{\gamma n^{4/3}}{\eta} \right)\left(\frac{1}{\rho} - 1 \right)^{2/3} R(t)^3 \right)}{\ln(1/\varepsilon)}$$

3-th stage of viscous sintering

Taking in account processes such are *densification* and *coarsening*, the fractal dimension is expected to slightly increase. In the limiting case of ideal sintering, $D_f \to 3$.

In fact, since the powder material is porous, there are two aspects of such a material fractality: the positive space made by grains collection and the negative space which is holes and pores collection. Ceramics materials during sintering process changes its inner morphology by shrinking pores and increasing its solidity and compatibility. But, after sintering it still contains residual pores, which means that any ceramics bulk CB can be represented as the positive space CB^+ and negative space CB^- union whilst these spaces are strictly separated. In formulas

$$CB = CB^+ \cup CB^- \quad \text{and} \quad CB^+ \cap CB^- = \varnothing .$$

This fact implies that if CB+ is a fractal set, its complement CB− is also a fractal set. During sintering process, the solidification takes part which includes bigger pores shrinking and disappearing of smaller ones. By using the known formulas for so called box counting method of evaluating fractal dimension it can be establish $DH_f(t)$ as a function of sintering time t, in three sintering phases. In fact, by the Frenkel formula for the sintering initial phase,

The pore radius R(t) is a function of time, with initial value R0 = R(0), σ is crystal lattice constant, η is the viscosity of the material, n is the number of pores per unit volume, ρ is the normalized density (normalized by the theoretical density), and γ is the surface energy.

Note that the pores' radii decrease as much as the elasticity module can permit it. Since this function is decreasing the above formulas predict that the limiting value for $DH_f(t)$ at the end of sintering process will be close to 3. Indeed, theoretically, $\lim_{t\to\infty} DH_f(t) = 3$.

Concerning these two subspaces CB+ -the ceramic material and CB$^-$ - pores collection, we came forward with a fact ([34]) that each subspace contributes by its own fractality. Namely, the morphology of grains, their fractal dimension in unity with intergranular contacts pattern introduce a "grain surface" factor, denoted by α_S. It is suitable to bound this dimensionless quantity so that $0 < \alpha_S < 1$. Consequently, the pores set also induce a pore-parameter $0 < \alpha_P < 1$, representing the ceramics body „negative space".

Arguing about the crystal surface "natural roughness" as macroscopic steps collection on the arbitrary section surface of the crystal plane section, the authors of [16] quote an observation Frenkel [17] 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. Such impact generates a motion inside the ceramics crystals in the Fermi gas form, containing different particles such as electrons (Bloch wave), atoms, atomic nuclei etc.

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16. L. E. Geguzin, N. I. Ovčarenko, Poverhnostnaya energiya i processy na poverhnosti tverd'h tel, Uspehi F. N. (1962) 76 (2), 283-324.

17. Ya. I. Frenkel, O poverhnostnom polzanii častic u kristallov i estestvennaya šerohovatosť estestvennyh granej, ŽETF 16 (1),(1948).

18. V.V. Mitić, V. Paunović, Lj. Kocić, Dielectric Properties of BaTiO3 Ceramics and Curie-Weiss and Modified Curie-Weiss Affected by Fractal Morphology, in: Advanced Processing and Manyfacturing Technologies for Nanostructured and Multifunctional Materials (T. Ohji, M. Singh and S. Mathur eds.), Ceramic Engi¬neering and Science Proceedings, Vol. 35 (6) 2014, pp. 123-133.

Dielectric constant of ferroelectrics materials depend on temperature and reaches a maximum value at Curie temperature and decreases with further increase in temperature according to the Curie-law $\varepsilon_r = C/T - T_o$. One of the reasons to use a modified $BaTiO_3$ is that, the additives have the effect of shifting Curie temperature i.e. shifts the maximum value of permittivity in the temperature range in which it can be exploited. The additives which have a higher valency then those being replaced, when present at level exceeding about 0.5wt% generaly inhibit crystal growth and move the Curie temperature towards lower values. Higher valency additives at low concentrations <0.2wt% generally lead to low resistivity of the modified $BaTiO_3$.

For the investigation ferroelectrics behavior in paraelectric phase beside the Curie Weiss law we can use modified Curie-Weiss relations which describe the deviation from linearity $\varepsilon_r = f$ (T) due to the diffuse phase transformations and the dielectric constant frequency dependence*.

The purpose of this paper is the dielectric properties Ho doped $BaTiO_3$ ceramics investigation in function of different amount of dopant concentration and fractal correction related to fractal-like structure of the material. The Curie-Weiss and modified Curie-Weiss law is used to clarify the influence of dopant on the dielectric properties and phase transformation in $BaTiO_3$.

* I. Isupov, " Some problems of diffuse ferroelectric phase transition", *Ferroelectric* 90, (1989).

The Curie temperature (T_C), determined from the maximum of the dielectric constant ε_r in the dielectric temperature characteristic, was in the range from 124 to 129^{o}C being lower for low doped Ho-$BaTiO_3$.

All specimens have almost sharp phase transition and follow the Curie-Weiss law (Fig. 4). For T > Tc (Tc- the Curie temperature), ε_r obeys the Curie-Weiss law

$$\varepsilon_r = \frac{C}{T - T_c}$$

with Curie constant C=1.5·10^5.

The Curie constant (C) decreases with the increase of additive amount and have an extrapolated Curie-Weiss temperature (T_0) down to lower temperature (Fig.5 and Fig.6). In 0.01wt % doped samples, that exhibit a high density, the Curie constant is higher compared to the high doped samples. It is known that the value of Curie constant is related to the grain size and porosity of samples. The highest value of C (C=1.50·10^5) was measured in 0.01 Ho-$BaTiO_3$-ceramics. The Curie constant C and the Curie-Weiss temperature T_0 values were given in Table 1.

In order to investigate the Curie-Weiss behavior, modified Curie-Weiss low is used. To quantify the diffuseness i.e. the diffuse phase transformation of ε_r at T_{max} the equation proposed by Uchino and Nomura* has been used:

$$\frac{1}{\varepsilon_r} - \frac{1}{\varepsilon_{r\max}} = \frac{(T - T_{\max})^{\gamma}}{C'}$$

were (γ) is the critical exponent of nonlinearity and C' is a Curie like constant.

The critical exponent gama (γ) was calculated from the best fit of curve $\ln(1/\varepsilon_r - 1/\varepsilon_m)$ vs. $\ln(T - T_m)$, where the γ represents the slope of curve (Fig 7).

The critical exponent γ is in the range $1 \leq \gamma \leq 2$, 1 for a sharp phase transformation and 2 for diffuse phase transformation. For BaTiO3 single crystal γ is 1.08 and for modified BaTiO3 gradually increases up to 2 for diffuse phase transformation.

* K. Uchino, S. Nomura; *Ferroelectric* Lett. Sec., 44, 55–61, (1982)

This temperature consideration illustrates impact on dynamical processes inside the ceramics body. Such impact generates a motion inside the ceramics crystals in the Fermi gas form, containing different particles such as electrons (Bloch wave), atoms, atomic nuclei etc. In essence this motion has Brownian character and impose necessity of introducing the third fractality factor–factor $\alpha_M \ (0 < \alpha_M < 1)$.

Our hypothesis is that $BaTiO_3$-ceramics working temperature must be influenced by these three fractality factors, making correction of „theoretic" temperature as

$$T_f = \alpha T,$$

where *a* is fractal corrective factor. It is natural that all three factors a_S, a_P, a_M, influences a as follows

$$\alpha = \Phi\left(\alpha_S, \alpha_P, \alpha_M\right)$$

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 aS, aP and aM, 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,

$$a = \frac{T_f}{T} = \frac{T - \mathrm{V}T}{T}$$

so that

$$0 < a = 1 - \mathrm{V}T / T < 1$$

Back to capacity formula gives us a hint of alpha correction embodied in the corrective coefficient a0 . To find connection between a0 and a consider the Curie-Weiss law, giving temperature dependence dielectric constants of BaTiO3-ceramics grains' contact zone

$$\varepsilon_r(T) = \frac{C_c}{T - T_S}$$

where T_S is the Curie temperature and C_c is Currie constant. If ε_r is corrected to gain the value $\alpha_0\varepsilon_r$, then from (5) and (7) follows $\alpha_0\varepsilon_r = C_c / (\alpha T - T_S)$. After elimination of Currie constant C_c using (7), one gets

$$\alpha = \frac{1}{\alpha_0} + \left(1 - \frac{1}{\alpha_0}\right)\frac{T_C}{T}, \text{ and reversely } \alpha_0 = \frac{T - T_C}{\alpha T - Tc},$$

which are the formulas connecting α and α_0 with temperature T. The Figure 6 visualize relationship between α and α_0, where the natural range for both α and α_0, are deliberately extended for better insight.

Consider now, an intergranular contact impedance as shown in Figure 7. Here, C_e is the main capacity component while C, R and L are parasite capacity, resistivity and inductivity resp., without α-correction, which means that intergranular geometry is considered as being flat. It is not difficult to see that the equivalent impedance, with α-correction included should be

$$Z_e = \frac{1 + \alpha_0\left(CRs + CLs^2\right)}{\alpha_0(C_e + C)s + \alpha_0^2 C_e C(R + Ls)s^2}, \quad s = j\omega = j2\pi f,$$

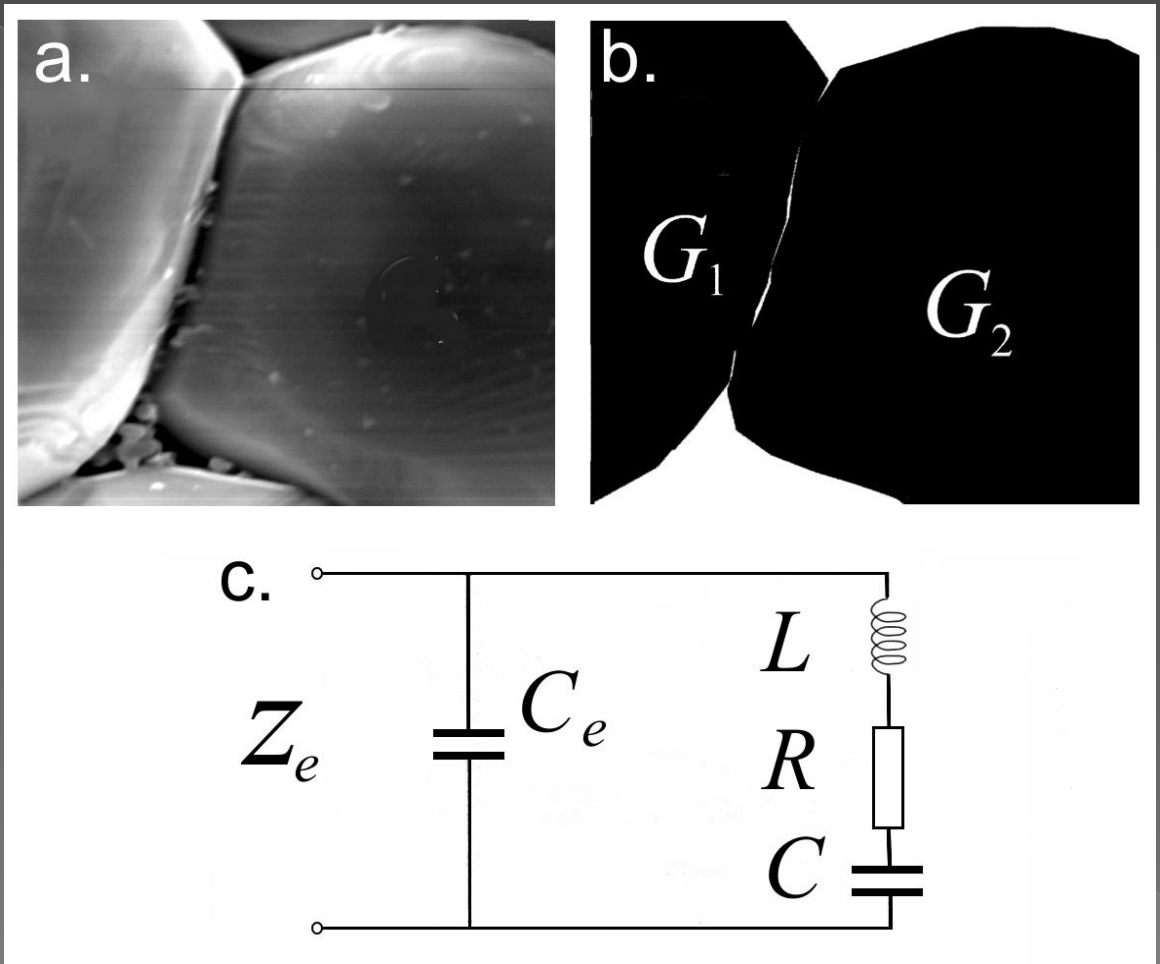


Fig. 6. a.Two grains of BaTiO3-ceramics doped with 0.1% of Ho_2O_3, sintered at 1350□C.
b. Shematic illustration of G_1-G_2 contact. **c.** Equivalent micro-impedance without *a*-correction.

or, expressed via frequency

$$Z_e = \sqrt{\frac{A^2}{4\pi f^2 B^2} + \frac{\alpha_0{}^4 C^4 R^2}{D^2}} \quad ,$$

where

$$A = 4\pi^2 \alpha_0{}^2 C^2 f^2 (4\pi^2 \alpha_0 C_e f^2 L^2 + \alpha_0 C_e R^2 - L) - 8\pi^2 \alpha_0{}^2 C C_e f^2 L + \alpha_0 vC + \alpha_0 C_e \ ,$$

$$B = \alpha_0 C^2 (4\pi^2 \alpha_0 C_e{}^2 f^2 (4\pi^2 f^2 L^2 + R^2) - 8\pi^2 \alpha_0 C_e f^2 L + 1) + 2\alpha^2 C C_e (1 - 4\pi^2 \alpha_0 C_e f^2 L) + \alpha^2 C_e{}^2 \ ,$$

$$D = 4\ \pi^2 \alpha_0{}^4 C^2 C_e{}^2 f^2 R^2 + (\alpha_0 C (4\ \pi^2 \alpha_0 C_e f^2 L - 1) - \alpha_0 C_e)^2 .$$

In this manner, Z_e includes, except C_e, C, L, R and f, also the corrective factor α_0 and, by help of formula (8), it also depends on α as it is shown by the graphs in Figure 8. Similar is shown in Figure 9.

If more than two grains are in contact, and this is the most common case in ceramics bulk, the situation becomes much more complicated. Let the simple case of four grains in contact be considered (Figure 10a). If these grains has four contacts, then the configuration of intergranular impedances will have form of a tetrahedron (Fig. 10b). The equivalent impedance between the grains G_1 and G_4, is the same as between points A and B on tetrahedral scheme. The triangles $Z_2Z_3Z_4$ and $Z_3Z_5Z_6$ can be transform in the corresponding "stars" $Z_aZ_bZ_c$ and $Z_pZ_qZ_r$ with

$$Z_a = \frac{Z_2 Z_4}{\Sigma'},\ Z_b = \frac{Z_2 Z_3}{\Sigma'},\ Z_c = \frac{Z_3 Z_4}{\Sigma'},$$
$$Z_p = \frac{Z_3 Z_5}{\Sigma''},\ Z_q = \frac{Z_3 Z_6}{\Sigma''},\ Z_r = \frac{Z_5 Z_6}{\Sigma''},$$

where $\Sigma' = Z_1 + Z_2 + Z_3$ and $\Sigma'' = Z_3 + Z_5 + Z_6$. The equivalent circuit is shown in Figure 11, and the final calculation gives

$$Z_{AB} = \frac{Z_1 (Z_a + Z_b)}{Z_1 + Z_a + Z_b} + \frac{(Z_b + Z_p)(Z_c + Z_q)}{Z_b + Z_c + Z_p + Z_q},$$

or, taking into account (10)

$$Z_{AB} = \frac{Z_1 Z_2 (Z_3 + Z_4)}{Z_1 (Z_1 + Z_2 + Z_3) + Z_2 (Z_3 + Z_4)} +$$
$$\frac{Z_3 \big(Z_6 (Z_1 + Z_2) + Z_3 (Z_4 + Z_6) + Z_4 (Z_5 + Z_6)\big)\big(Z_5 (Z_1 + Z_3) + Z_2 (Z_3 + 2Z_5 + Z_6)\big)}{(Z_1 + Z_2 + Z_3)(Z_3 + Z_5 + Z_6)\big((Z_1 + Z_4)(Z_5 + Z_6) + Z_3 (Z_4 + Z_5 + Z_6) + Z_2 \big(Z_3 + 2(Z_5 + Z_6)\big)\big)}.$$

Suppose the all the impedances are equal to some Z. Then, according to (11), $Z_{AB} = \frac{5}{3} Z$.

If the parasite components L, R and C in the circuit at (9) are neglected, and if $1/Z_i = j\omega\alpha_0 C_e = j 2\pi f\ \alpha_0 C_e$, all i , then

$$Z_{AB} = \frac{5}{3} \frac{1}{j\,\omega\,\alpha_0 C_e} = \frac{1}{j\,\omega (3\alpha_0 C_e / 5)}.$$

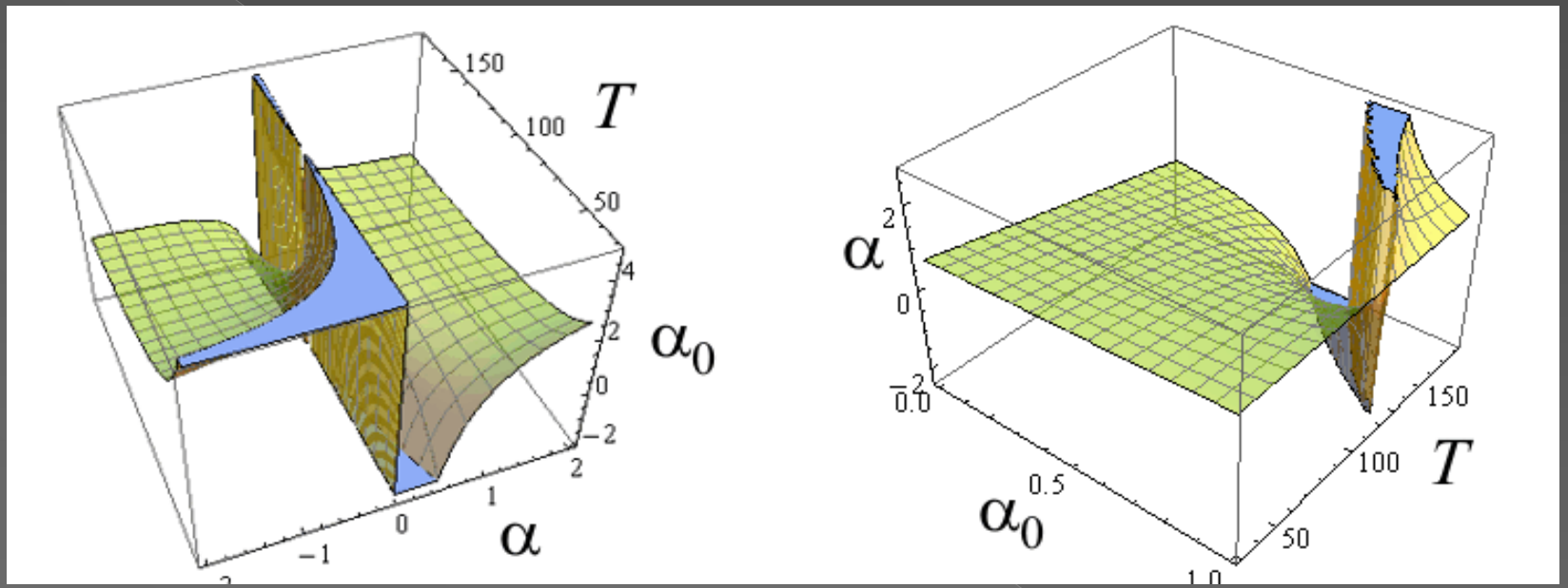


Fig. 7. Left. c_e=0.1; c=0.01; L=0.001; Right. c_e=0.01; R=1; L=0.001;{a,0,1},{f,1,80}.

Suppose the all the impedances are equal to some Z. Then, according to (11), $Z_{AB} = \frac{5}{3} Z$.

If the parasite components L, R and C in the circuit at (9) are neglected, and if $1/Z_i = j\omega\alpha_0 C_e = j\,2\pi f\ \alpha_0 C_e$, all i , then

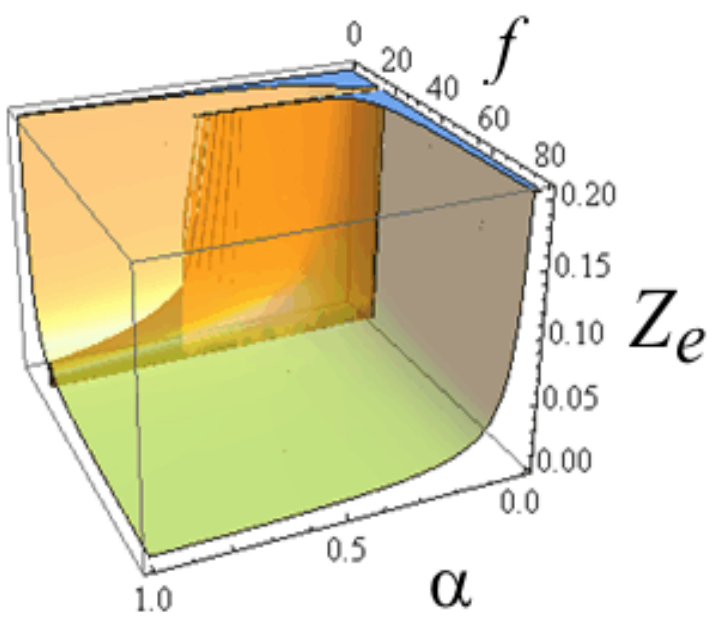
$$Z_{AB} = \frac{5}{3}\frac{1}{j\,\omega\,\alpha_0 C_e} = \frac{1}{j\,\omega\left(3\,\alpha_0 C_e / 5\right)}.$$

This formula reveals that, the overall capacity in the case of tetrahedral configuration is smaller than the single contact capacity, $C_{AB} = (3/5)\ \alpha_0\ C_e$.

Similar calculations in the case of eight grains in contact, arranged in cubic manner (Figure 11, right), gives

$$Z_{AB(\mathrm{CUBE})} = \frac{5}{6}\frac{1}{j\,\omega\,\alpha_0 C_e} = \frac{1}{j\,\omega\left(6\,\alpha_0 C_e / 5\right)},$$

and therefore, $C_{AB} = (6/5)\ \alpha_0\ C_e$, so the capacity of the cubic cluster is bigger than the capacity of a single contact.



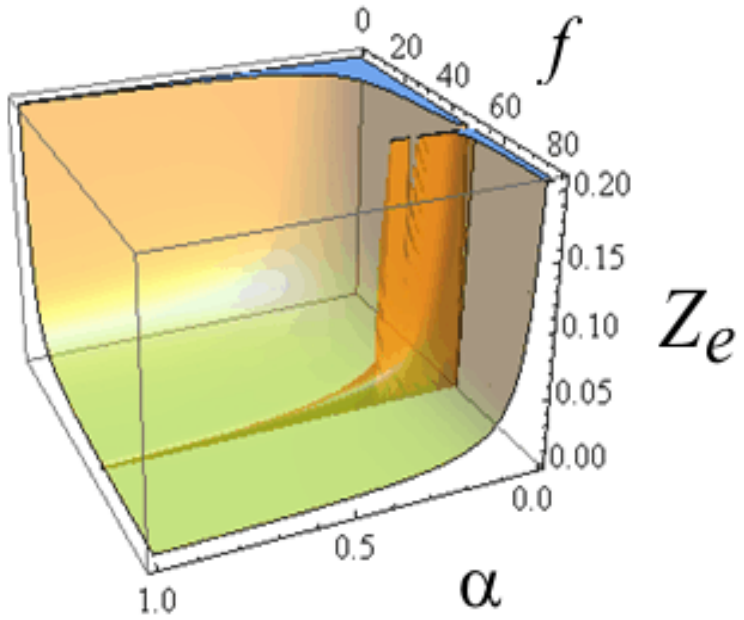


Fig. 8. The intergranular impendence Z_e given by (9) as a function of $0 < a < 1$ and $5 < f < 50$, T=80°C for c_e=0.5; c=0.01; L=0.01; R=0.002 (left); and for c_e=0.5; c=0.001; L=0.01; R=0.002; (right). Physical units are neglected.

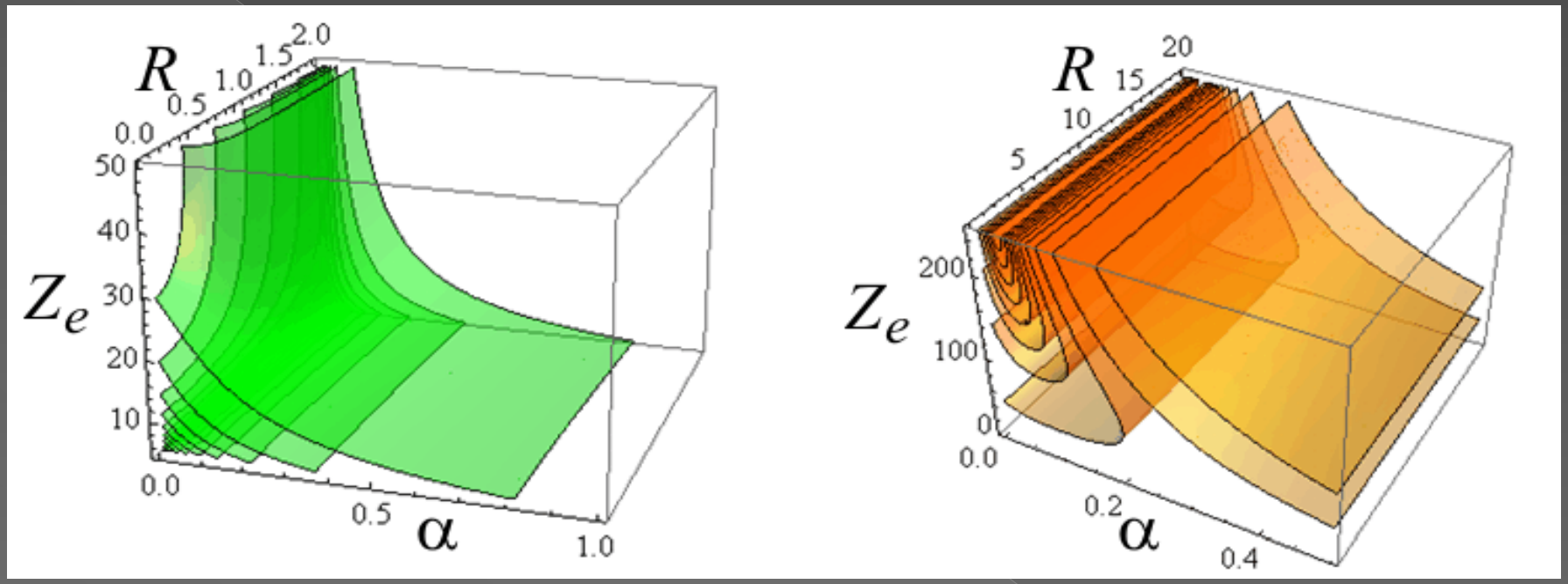


Fig. 9. The level surfaces of intergranular impendence Z_e given by (9) as a function of $0 < a < 1$, $0 < R < 2$, T=80°C and f = 1(8)80 and for c_e=0.1; c=0.01; L=0.001 (left); c_e=0.01; R=1; L=0.001 (right); Physical units are neglected.

If more than two grains are in contact, and this is the most common case in ceramics bulk, the situation becomes much more complicated. Let the simple case of four grains in contact be considered (Figure 10a). If these grains has four contacts, then the configuration of intergranular impedances will have form of a tetrahedron (Fig. 10b). The equivalent impedance between the grains G_1 and G_4, is the same as between points A and B on tetrahedral scheme. The triangles $Z_2Z_3Z_4$ and $Z_3Z_5Z_6$ can be transform in the corresponding “stars” $Z_aZ_bZ_c$ and $Z_pZ_qZ_r$ with

$$Z_a = \frac{Z_2 Z_4}{\Sigma'},\ Z_b = \frac{Z_2 Z_3}{\Sigma'},\ Z_c = \frac{Z_3 Z_4}{\Sigma'},$$
$$Z_p = \frac{Z_3 Z_5}{\Sigma''},\ Z_q = \frac{Z_3 Z_6}{\Sigma''},\ Z_r = \frac{Z_5 Z_6}{\Sigma''}, \qquad (10)$$

$$\Sigma' = Z_1 + Z_2 + Z_3$$

$$\Sigma'' = Z_3 + Z_5 + Z_6$$

The equivalent circuit is shown in Figure 11, and the final calculation gives

$$Z_{AB} = \frac{Z_1\left(Z_a + Z_b\right)}{Z_1 + Z_a + Z_b} + \frac{\left(Z_b + Z_p\right)\left(Z_c + Z_q\right)}{Z_b + Z_c + Z_p + Z_q},$$

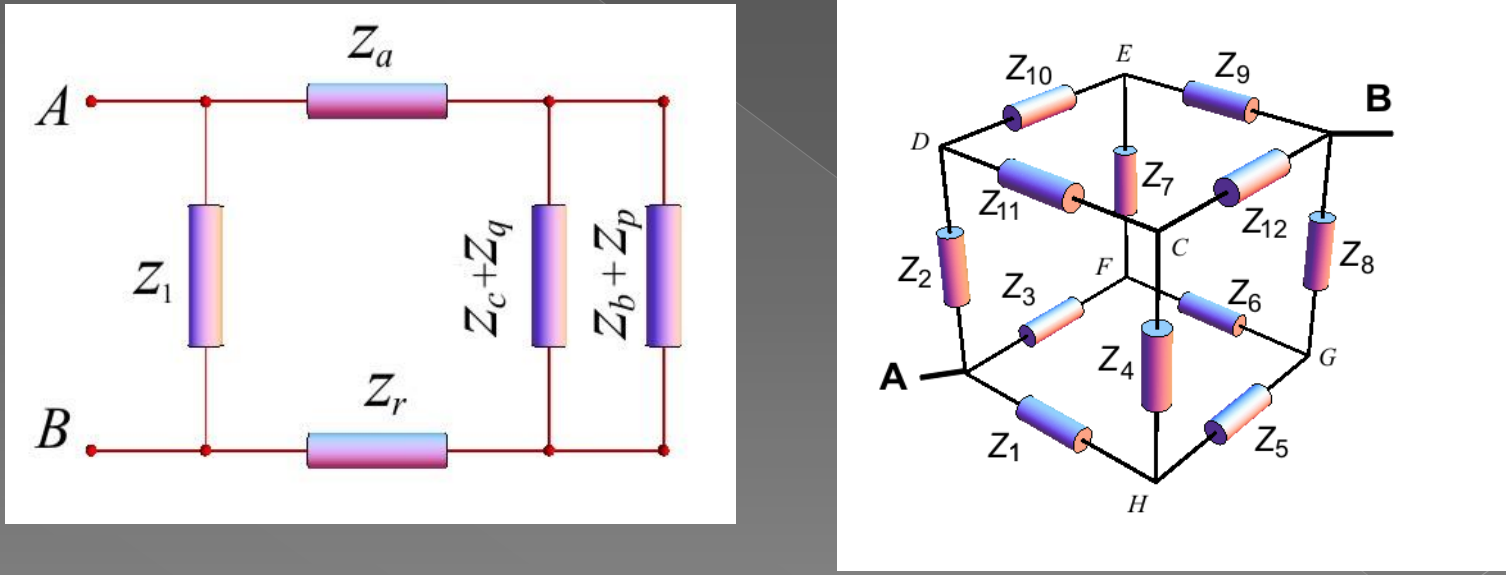


Fig. 10. Left. The equivalent circuit for the four grains contacts. Right. Cube of impedances.

or, taking into account (10), one gets

$$Z_{AB} = \frac{Z_1 Z_2 (Z_3 + Z_4)}{Z_1 (Z_1 + Z_2 + Z_3) + Z_2 (Z_3 + Z_4)} + \frac{Z_3 \left(Z_6 (Z_1 + Z_2) + Z_3 (Z_4 + Z_6) + Z_4 (Z_5 + Z_6)\right)\left(Z_5 (Z_1 + Z_3) + Z_2 (Z_3 + 2Z_5 + Z_6)\right)}{(Z_1 + Z_2 + Z_3)(Z_3 + Z_5 + Z_6)\left((Z_1 + Z_4)(Z_5 + Z_6) + Z_3 (Z_4 + Z_5 + Z_6) + Z_2 (Z_3 + 2(Z_5 + Z_6))\right)}.$$

Suppose the all the impedances are equal to some *Z*. Then, according to (11), $Z_{AB} = \frac{5}{3} Z$. If the parasite components *L*, *R* and *C* in the circuit at (9) are neglected, and if $1/Z_i = j\omega\alpha_0 C_e = j2\pi f\ \alpha_0 C_e$, all *i*, then

$$Z_{AB} = \frac{5}{3} \frac{1}{j\omega\ \alpha_0 C_e} = \frac{1}{j\omega(3\alpha_0 C_e / 5)}$$

This formula reveals that, the overall capacity in the case of tetrahedral configuration is smaller than the single contact capacity,

$$C_{AB} = (3/5)a_0\ Ce.$$

Similar calculations in the case of eight grains in contact, arranged in cubic manner (Figure 11, right), gives

$$Z_{AB(\mathrm{CUBE})} = \frac{5}{6}\frac{1}{j\omega\ \alpha_0 C_e} = \frac{1}{j\omega\left(6\alpha_0 C_e / 5\right)}$$

and therefore, $C_{AB} = (6/5)\ \alpha_0\ Ce$, so the capacity of the cubic cluster is bigger than the capacity of a single contact.

OUTLOOK

In this article the doped $BaTiO_3$-ceramics fractality and some consequences are investigated. It is not new that ceramics, from powder phase through all sintering phases, exhibits fractal structure and microstructure posting the basis for ceramics' dielectric, ferroelectric, PTCR and piezoelectric properties. Based on previous investigations (papers [7-35]) where some of $BaTiO_3$-ceramics elements fractality and also for doped $BaTiO_3$-ceramics were established, a new approach to intergranular capacity is developed. Also, the relationship between the size of contact area and its fractal dimension is formulated. It is shown that the fractal form of an intergranular contact zone may be presented as a chain of micro-capacitors forming one bigger capacitor. It is shown how the capacity increases as the complexity of contact zone increases. The *alpha correction* of intergranular capacity is introduced to reflect increasing of capacity due fractal character of intergranular contact. The correction factor is a0>1 Next, the pores geometry in $BaTiO_3$-ceramics material is investigated with conclusion that all pores collection in ceramic body is a fractal object too. The porosity behavior is studied in all of sintering process three phases, Frenkel, Scherer and Mackenzie-Shuttleworth, and corresponding formulas for box-dimension of pores are established. Third source of fractality, except intergranular surface geometry quantified by the factor aS and pore system that yields factor aP, in $BaTiO_3$-ceramics is interior movements of different particles, subatomic, atomic, molecular etc. Due to its Brownian nature, it also has fractal character defined by the third factor aM. These three factors aS , aP, and aM, are arguments of a functional parameter a that represents all of them. The relationship between a and a0 is established and two different models of grain clustering is considered. Four grains-tetrahedral, that diminishes total capacity of four intergranular contacts for 40% while the eight grains-cubic connection increases it for 20%.

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