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Fractal nature structure reconstruction method in designing microstructure properties

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ABSTRACT

In the process of predicting materials properties, it is necessary to predict microstructures. This is important for ceramic materials, for the purpose of miniaturization and for a higher level of integration. Although the method of material structure reconstruction is for the first time used on $BaTiO_3$-ceramics by these authors, it can also be used for silicate, refractory and other ceramics, and, for any powder-based material. The reconstruction is based on the grain perimeter fractal analysis and the Richardson method. The main contribution of this paper is establishing the relationship between the electronic properties of $BaTiO_3$-ceramics and its microstructural fractal nature. Similar relations can be used for any ceramics or even any materials. The samples consolidation includes both powder pressing (cold sintering phase) and sintering in a furnace (hot sintering phase). The fractal nature characterization has a very important role from the powder phase up to the final microstructure, through which structures it directly impacts electro-physical and other ceramics properties. Thereby, the new possibilities in the microstructure characterization applications are directly introduced to an engineering system, providing the new solutions related to computer aided support. This creates new industrial production and application possibilities, especially in the process of ceramics material consolidation, now based on microstructural properties, which is providing a basis for "fractal electronics" development.

1. Introduction

The ability to predict microstructures and describe them mathematically is essential for material-property prediction. The goal of this paper is to achieve a certain structure and final material's properties, within the material's consolidation triad: Synthesis – Structure – Properties. Already known microstructure characterization analysis methods, aside from the parameter characterization, do not offer possibilities for demonstrating grain shape reconstruction. Once reconstructed, the SEM materials characterization data do not play an active role in the microstructure shapes for the purpose of microstructure property prediction. All the available microstructure analysis tools are merely the passive instruments for obtaining the characterization data. So, the fractal nature structure analysis is practically revitalizing all of these known methods, making them applicable for emerging future needs, especially in the microelectronics miniaturization area [1,2]. There are several similar options in scientific/technological processes expressed in different scientific terms and options., The research results related to them found in the scientific literature include: material's properties programming, properties prediction, designing, material's programming, and, (the latest) – new simple material's modeling (NSMM).

All the most modern and maximally optimized microstructure methods face the question: how much flexibility can be provided regarding the reconstruction of the structural units (grains and pores) and improvement of higher miniaturization levels? They are in favor of microelectronic components and devices, as well as, electronic circuit integration. Herewith, new frontiers based on the fractal nature of materials structure opens up. Three points are common: (i) much deeper material characterization; (ii) reconstruction of the structure units and (iii) prediction of the microstructure properties within the miniaturization framework.

The grain and pore prediction methods based on the measurements of the solid Euclidean geometric statistical characteristics are given in [3–9]. In a preferred scope, the methods almost always involve a microscopic examination of the number of geometric features of the grain and pore surface under several levels of magnification. The main characteristic of any fractal object A is its fractal dimension *DH* (which

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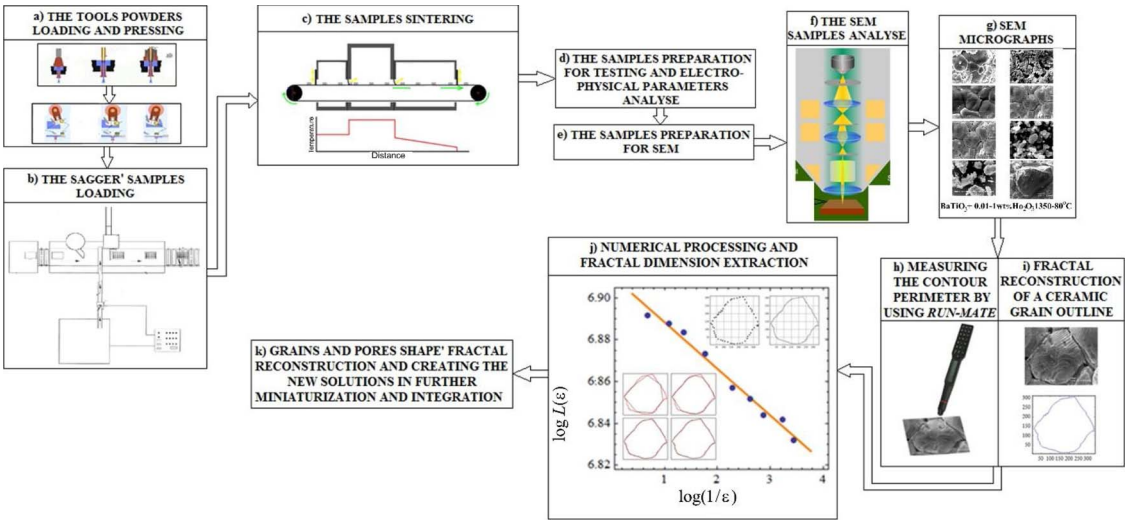

Fig. 1. Experimental procedure.

is a real number, smaller than the geometric dimension of the minimal space containing A). The authors' standpoint is that the fractal dimension of the grains' or pores' space considerably contributes to the physical-chemical properties of ceramics and other structures. In the last 20 years, there has been a boom of a wide range of applications of fractals in different disciplines [10–15]. The generalized fractal phenomena were discovered in physical systems [16] but also applied to environmental protection [17]. For ideal (mathematical) fractals, the *DH* is independent on the measure scale. But, natural fractals, like clouds, trees, tempestuous sea surface, dust in the atmosphere, interstellar dust grains, the solar spectrum, ore distribution in the Earth's crust, pollution of waters, heart oscillations, yearly variations in insect population; and ceramics are *almost* fractals. This means that ceramic structures are much closer to ideal fractals than to any Euclidean object. But, these are not self-similar structures as the majority of mathematical fractals (Cantor set, Sierpinski Triangle etc.). On the other hand, many fractals (Mandelbrot, Julia sets etc.) also do not conform to self-similarity. Instead, they are statistically self-similar, which is a kind of a "self-similarity", characteristic of all "natural fractals".

By using measured data, such properties as the porosity, electrical conductivity, and permeability can be expressed in terms of fractal dimensions. The fractal methods may be used to determine grain and pore dependent electro-physical properties of various microporous solids, including reservoir rock, heterogeneous catalyst materials, and electrochemical electrodes. These methods may be used automatically employing a specially designed measurement system comprised of a microscope or a microdensitometer and a signal processing devices including a computer.

It was only natural to step up the research in the fractal nature of ceramics and similar materials with a special focus on their fractal grainy and porous microstructures [18] and micro-capacity properties [19–21]. The issues of micro-capacity and integral capacity are of great importance since the collection of a vast number of micro-capacitors can be an energy storage.

It is known that the intergranular contacts inside the $BaTiO_3$-ceramic materials are responsible for intergranular capacity and, specifically, for the phenomena of high capacity realized with barium-titanate ceramics and ceramics based on the other perovskite materials. This phenomenon is quantified in [22] by introducing a *fractal correction factor α*. It is preconceived as a multiplicative constant for the permittivity ε_r of a specific ceramics material. In this manner, instead of ε_r in all relevant formulas we set $\alpha\varepsilon_r$, where α is a dimensionless real number that compensates for the increasing capacity that comes from fractal nature of the intergranular contacts. One way to estimate the value of α is to extract it out of the fractal dimension *DH* of the outline or the surface of ceramic grains. In the meantime, this idea was further elaborated before it assumed its final form.

Once α is estimated for a specific kind of ceramics, the introduction of the temperature variation via the Curie-Weiss law is facilitated, as will be shown below. In this way, the fundamental thermodynamic regime inside the ceramic body is influenced by parameter α. For the scope of developing the idea of fractal nature of ceramic materials, see [23–25].

The experiments are based on several taken SEM micro photographs of different specimens. In fact, since 1995, when our group started studying the fractal aspect of different ceramic materials microstructure, a certain number of papers devoted to this issue was based on several thousand of specimens with or without additives.

The "fractal nature structure analysis – fractal electronics" relation, practically, offers a new scientific approach, closely followed by nanoscience and nanotechnologies.

2. Experimental procedure

In this paper, we used $BaTiO_3$-ceramics for the specific fractal application and it was necessary to prepare the sample series based on $BaTiO_3$-ceramics for further research related to optimizing the interdependence of microstructural and electrical characteristics. The effects of pressing pressure, sintering conditions (temperature, sintering time) and the additives' content on the microstructure and thus on the final $BaTiO_3$-ceramics characteristics, were investigated [22,25].

For this particular research, pure and Ho doped $BaTiO_3$ceramics, from 0.01 wt% to 1 wt%, of Ho were used. The complete experimental procedure is shown in Fig. 1. After measuring the pure and doped BaTiO3 powders, the mixture was processed in a ball mill. Homogenization was carried out for about 48 h. After milling, the powders were dried for several hours. Then, the material was collected in a special vessel and its bulk density was checked every hour. The obtained samples were double-sided pressed with pressures ranging from 98 to 150 MPa. The double-sided pressing provides a greater homogeneity of investigated samples. The $BaTiO_3$ sample pressing is done on a hydraulic press (JAPAN - KYOTO Murata MFG - TYPE - FMC - 1-Kilkis) (Fig. 1a).

Sintering was performed in an electric tunnel furnace (CT-10 Murata) at the temperatures ranging from 1290 °C to 1380 °C (Fig. 1c). These samples were placed in special containers (saggers) for 38 h of the entire process. Prior to sintering, the samples had been covered with refractory special powder sand in order to be separated easily after the