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Automatic Microstructure Analysis of Sintered Materials

In this article the new approach to automatic microstructure analysis of the sintered materials has been presented. The method is based on contour recognition and decomposition of digital image, according to their gray scale intensity and the use of log-hyperbolic grain size distribution and it has been succesfully applied on microstructure analysis of $BaTiO_3$ sintered under non isothermal conditions.

Keywords: *microstructure analysis, log-hyperbolic distribution, sintering*

1. INTRODUCTION

It is well known that microscopy-based techniques provide a powerful tool for qualitative and quantitative microstructure analysis of advanced sintered materials, especially for the analysis of their grain and pore sizes, their distribution and morphology [1]. The general procedures related to microscopy-based microstructure analysis include manual, semiautomatic and automatic techniques for quantitative analysis of the microstructure constituents [2]. Manual techniques of the microstructure analysis are often based on the use of a marking device such as a cross-hair or a graticule with inscribed circles of varying diameters. Semi-automatic techniques require human intervention, but offer, to some degree, the ease of automatic analysis. Some of these techniques are based on the area analysis, where grain sizes are determined from cross sectional area of the grain, while others are based on the image shear techniques. All these techniques reduce the appearance of the random errors which are often associated with manual counting and analysis. Automatic techniques for the microstructure analysis use a combination of the image capture and viewing techniques, followed by image interpretation using algorithms based mostly on Fourier and fractal shape analysis. The advantage of these techniques is in ability to use the computers and powerful algorithms in order to reduce the amount of the time required for the analysis.

In this article, a new approach to the automatic quantitative microstructure analysis is proposed and described. It basically includes two steps. The first step assumes the application of digital pattern recognition (DPR) method, which we have recently developed in order to analyse digital micrograph of a representative surface of a sintered material [3]. In the second step log-hyperbolic function is used for fitting the grain sizes (i.e. equivalent diameters) distributions, provided by DPR method. It should be mentioned that we are the first who have succesfully applied this type of function for the analysis of microstructure development of sintered materials and that some of the results of these analysis are presented in this article

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1.1. Digital pattern recognition method

Digital pattern recognition method is based on laws of photometry (automatic CMYK, gray scale and the analysis of the isohele contours for isolated and overlapped objects). In general, it can be divided into three main processes.

The first process in digital pattern recognition of digital images is determination of a pseudo-rectangles (3D (2D + intensity or density) polygons with 4 vertexes) in 3 views (screen, image and physical coordinates of microstructure) [4 - 7]:

1. The determination of referent positions of polygons (polygon vertexes) on screen and creation of a set of screen coordinates of polygons;
2. The calculation of the image coordinates of polygons by using file format description - I/O procedures [4, 8];
3. The input of the physical coordinates of polygons of the presented image.

The use of library functions (for solving linear and nonlinear algebraic equations) makes possible the elimination of deformities of images in the calculation process.

The second process in this method includes determination of the gray scale histogram of image as well as primary things-in-pattern detections (contours based on contrast-edge detection) [5, 8].

The third process is determination of the digitalization area and the determination of the gray scale resolution in decomposition of the image by the user.

It should be noticed that the multi layer (skin) analysis of the image and the 3D reconstruction of pictures (depth analysis) including overlay effects, can be also used in this process [4 - 6, 8]. The final process

is the acquisition of data using I/O processing, creating sets of similar patterns whose present reflection isohels of 3D objects, surface-volume analysis and the recalculation of digitized data in physical coordinates [5 - 8]. Due to these processes, the relative estimated errors of volume diameter distribution analysis is less than 1% [6, 7].

1.2. The mathematical basics of the log-hyperbolic distribution

It is widely recognized that empirical statistical distributions, even in the cases related to large samples, sometimes may signifficantly deviate from the most commonly used normal Gaussian distribution. Numerous researchers have invested respectable efforts to resolve this problem by applying different less or more complex mathematical models. Among others, a possible approach to overcome these situations is to employ the log-hyperbolic function (fig 1.)

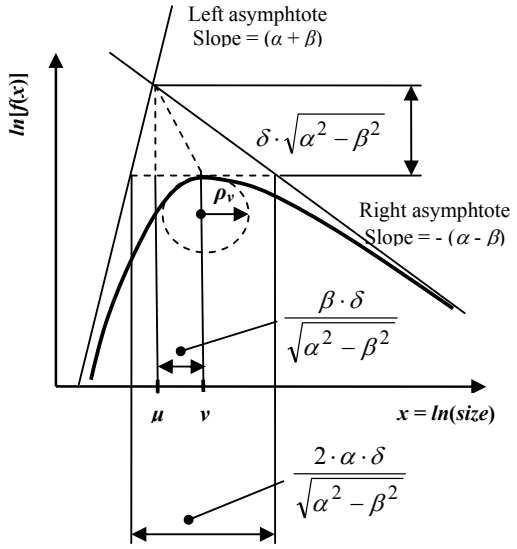


Figure 1. The log-hyperbolic function

This function was first introduced by Barndorf-Nielsen [9] to describe the asymmetric probability density distribution of the wind-blown sands particle sizes. Altough the applicabillity of this function in a wide variety of statistical problems related to geology, astronomy, biology etc. has been also reviewed [10,11], there is no report of the application of this function for calculation the size distribution of the characteristic diameter of sintered material grains.

In order to calculate the size distribution of the characteristic diameter of sintered material grains, in this paper we followed the numerical algorithm described by Bhatia and Durst [12]. It is based on the one-dimensional hyperbolic probability density function, which may be represented in the form:

$$f(x) = A(\alpha,\beta,\delta)\cdot e^{\left(-\alpha\sqrt{\delta^2+(x-\mu)^2}+\beta\cdot(x-\mu)\right)} \qquad (1)$$

The four arbitrary parameters: $\alpha > 0$, $|\beta| < \alpha$, $\delta > 0$ and $\mu \in (-\infty,\infty)$ that define its shape are evaluated numerically on the base of adequate empirical data set.

A is the constant, which enables the hyperbolic function (1) to satisfy the standard condition of the theory of probabilty:

$$\int_{-\infty}^{\infty} f(x)\cdot dx = 1 \qquad (2)$$

It is defined as:

$$A(\alpha,\beta,\delta) = \frac{\sqrt{\alpha^2-\beta^2}}{2\cdot\alpha\cdot\delta\cdot K_1\left(\delta\cdot\sqrt{\alpha^2-\beta^2}\right)} \qquad (3)$$

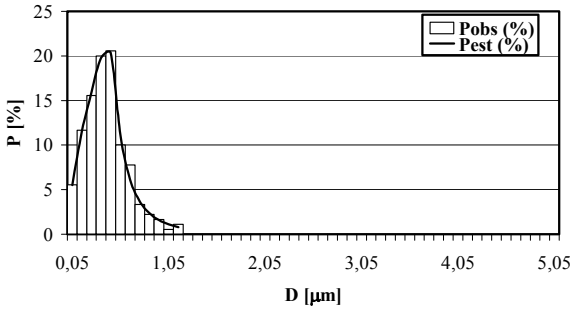
where $K_1(\cdot)$ is the Bessel function of the third kind and first order. If eq. (1) is applied on the logarithm values of size, instead of the variable x directly, the log-hyperbolic function arises (fig. 1).

2. EXPERIMENTAL PROCEDURE

Taking into account that $BaTiO_3$ is a multifunctional material, whose microstructure greatly influences its electrical properties, and that in many cases it can be used as a model material for sintering studies, in this article microstructure analysis of $BaTiO_3$ sintered under non isothermal conditions has been performed using DPR method. As the starting material, high purity commercial $BaTiO_3$ powder (Merck 99.8% Ba/Ti ratio of 1:1) was used. The binder free powders were compacted using the uniaxial double action pressing process in a 6 mm diameter tool (Hydraulic press RING, P-14, VEB THURINGER). Compacts were placed in an alumina boat and heated in a tube furnace (Lenton Thermal Design Type 1600). The samples were sintered under non isothermal conditions up to 1200°C, 1300°C, and 1380°C, with a heating rate of 10°C/min. Microstructure investigations of the samples were performed using a JEOL-JSM-T20 scanning electron microscope. This type of electron microscope enables observation of the samples surface by enlaging it to 35,000 times, with the resolution of 4.5 nm. The grain size and morphology were analysed by digital pattern recognition (DPR) microstructure quantity analysis and log-hyperbolic distribution model. Results are presented in figs. 2.a - 5.a, where P_{obs} denotes the size distribution histograms of the characteristic grain diameter, observed by DPR analysis, while P_{est} expressed adequate log-hyperbolic fit values.

3. RESULTS AND DISCUSSION

Microstructure analysis showed that the unimodal size distribution, characteristic for the starting material changed into multimodal distribution for the sintered samples. It should be noticed that mathematical model based on log-hyperbolic function, as well as most of available distribution models, supports the unimodal distribution shape. In our case this problem is resolved by partial fitting of the histogram related to the sintered material. The grain diameters range was divided into three subranges in the cases of materials sintered at 1200°C and 1300°C, and into two subintervals for the material sintered at 1380°C. Final agreement is good, as can be seen in figs. 2 - 5.



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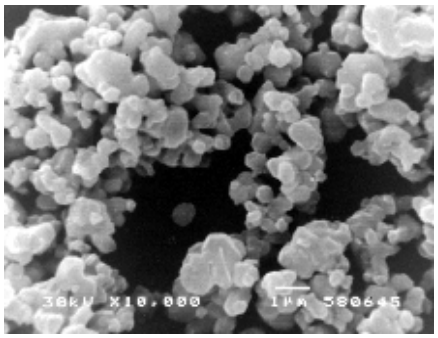
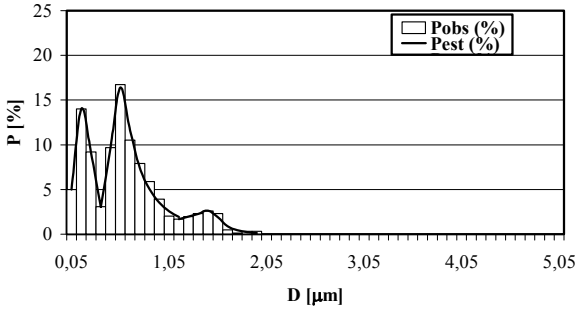


b)

Figure 2. Microstructure analysis of the initial powder: a) relative frequency versus grain diameter size, b) micrograph of the analysed sample.



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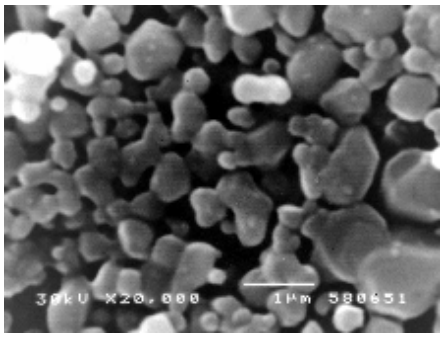

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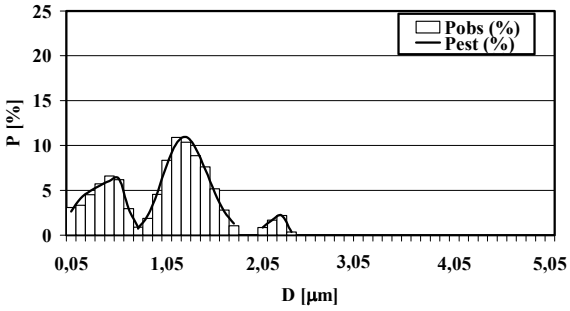
Figure 3. Microstructure analysis of the samples sintered at 1200°C: a) relative frequency versus grain diameter size, b) micrograph of the analysed sample.



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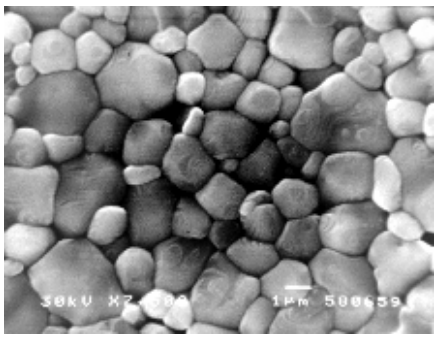
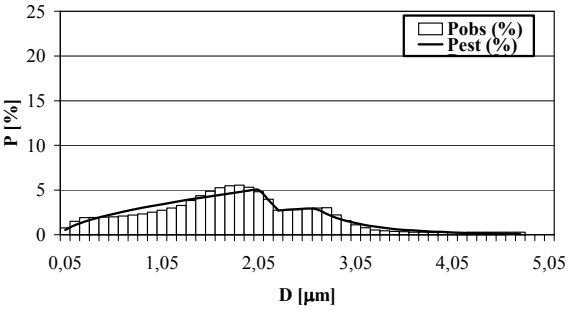


b)

Figure 4. Microstructure analysis of the samples sintered at 1300°C: a) relative frequency versus grain diameter size, b) micrograph of the analysed sample.



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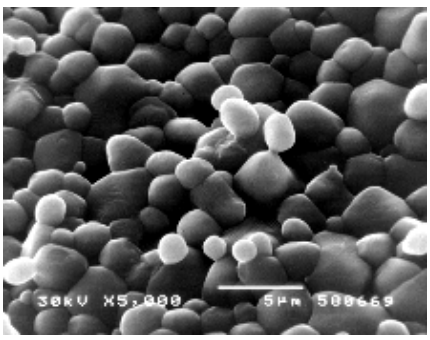

b)

Figure 5. Microstructure analysis of the samples sintered at 1380°C: a) relative frequency versus grain diameter size, b) micrograph of the analysed sample.

It is well known that the sintering process is generally the result of atomic motion, stimulated by the high temperatures. The driving force of the process is the reduction in the free energy by the reduction of the total surface energy of the system [13]. As a result, mass transport by plastic flow, bulk diffusion, surface and grain boundary diffusion, or a combination of these processes occurs, resulting in the increase of the grain size of the sample [14]. Since the material transport takes place, the sintering process can be divided into a number of stages such as: initial bonding among particles, neck growth, pore channel closure, pore rounding, shrinkage, grain growth.

According to our analysis, we have concluded that obtained types of distributions are characteristic for the morphologies at the early and medium stages of the sintering, during which formation of contact necks between grains and material transport occurs (fig. 3). During these stages, interaction between pores and grain boundaries may take some of the following forms: some pores can be dragged by moving of grain boundaries, resulting in the retardation of the grain growth, while some grain boundaries may brake away from the pores, leaving them isolated in the grain interior. Therefore, since the separation of pores from the boundaries limits the final sintered density, it is important to optimise grain boundary conditions by careful temperature control during sintering.

As the sintering temperature rises up to 1300°C and 1380°C, enhanced surface reactivity leads to the intensification of the transport processes resulting in a less porous and more dense microstructure, characterized by the grain size distribution showed in figs. 4 and 5. These types of distributions are characteristic for more advanced stages of sintering, when homogenisation of initial microstructure occurs. As a result of the mass transport between the grains which occur at these temperatures, we have noticed that the grains with a higher number of boundaries grow, while grains with a smaller number of boundaries decrease (fig. 6).

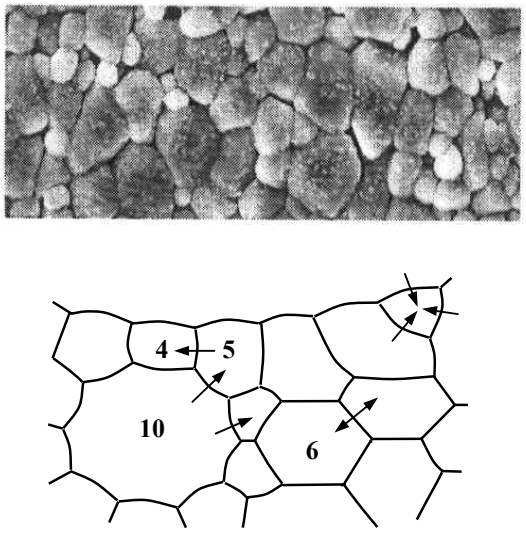

Figure 6. Grain boundary migration of sintered $BaTiO_3$

Our analysis showed that since the grain boundaries migrate towards their centers of curvature, grains with fewer than six sides tend to shrink, and those with more than six sides tend to grow.

Moreover, when the ratio of the grain radius toward the pore's curvature radius decreases to the zero, the interfaces are flat and have no tendency to shrink. But when the mentioned ratio is negative the pores tend to grow. Thus from a relationship between the number of surrounding grains and pore diameter to grain diameter ratio, one can derive conditions for microstructure constituents stability. These conclusions are very important since they can be related to specific properties of sintered materials and used for optimization of the shape forming conditions.

4. CONCLUSION

In this article, a new possible approach for the microstructure analysis of sintered material is presented. It includes an automatic contour recognition and decomposition of digital image of sintered material, according to their gray scale intensity and the use of log-hyperbolic function for fitting the resulting grain sizes. In order to analyse digital micrograph of a representative surface of sintered materials, the authors have developed and used digital pattern recognition (DPR) method. The resulting size distribution histograms of the characteristic diameter of sintered material grains are modeled by log-hyperbolic function. This new approach for the microstructure analysis has been succesfully applied to microstructure analysis of $BaTiO_3$ sintered under non-isothermal conditions. The change of the size distributions for various sintering temperatures has been explained according to sintering stages. The new approach for the automatic microstructure analysis can be used, either for quality control, in order to optimize the conditions of shape forming and densification, or in research, to give special parameters related to specific properties of sintered materials.

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АУТОМАТСКА МИКРОСТРУКТУРНА АНАЛИЗА СИНТЕРОВАНИХ МАТЕРИЈАЛА

Вера Павловић, Драган Петровић, Зоран Николић, Владимир Павловић

У овом раду је презентована нова метода за аутоматску микроструктурну анализу материјала. Метода је базирана на препознавању контура и декомпозицији дигиталне слике на основу фотометријских закона, као и на примени log-хиперболичне дистрибуције. Ова метода је успешно примењена за микроструктурну анализу неизотермски синтерованог баријум-титаната.