SINTERING TEMPERATURE INFLUENCE ON GRAINS FUNCTION DISTRIBUTION BY NEURAL NETWORK APPLICATION

by

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Artificial neural networks application in science and techonology begun during 20th century. This biophysical and biomimetic phenomena is based on extensive research which have led to understanding how neural as a living organism nerve system basic element processes signals by a simple algorithm. The input signals are massively parallel processed, and the output presents the superposition of all parallel processed signals. Artificial neural networks which are based on these principles are useful for solving various problems as pattern recognition, clustering, functional optimization. This research analyzed thermophysical parameters at samples based on Murata powders and consolidated by sintering process. Among different physical properties we applied out neural network approach on grain sizes distribution as a function of sintering temperature, T, (from 1190-1370 °C). In this paper, we continue to apply neural networks to prognose structural and thermophysical parameters. For consolidation sintering process is very important to prognose and design many parameters but especially thermal like temperature, to avoid long and even wrong experiments which are wasting the time and materials and energy as well. By this artificial neural networks method we indeed provide the most efficient procedure in projecting the mentioned parameters and provide successful ceramics samples production. This is very helpful in prediction and designing the micro-structure parameters important for advance microelectronic further miniaturization development. This is a quite original novelty for real micro-structure projecting especially on the phenomena within the thin films coating around the grains what opens new prospective in advance fractal microelectronics.

Key words*: neural networks, sintering temperature, fractal microelectronics, micro-structure miniaturization, biomimetic*

Introduction

During last few decades many non-conventional methods are applied solving engineering problems. They imply usage of fuzzy and genetic algorithms as well as neural net-

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works. Lately, combination of these methods named hybrid intelligent approach has a widespread application. This approach is used solving various fields such as: medicine [1], geology [2, 3], energetics [4, 5]. Having this in mind we decided to investigate possible neural networks application on sintering process.

The main idea in this paper is to extend the neural network application on processes in sintering within different sintering temperatures intervals. The consolidation process of ceramic materials at different thermal conditions has a very important relation to sintering temperature. First, in this research, we introduce the review of some results based on neural networks applied for different physical parameters. Then we review the results in some our previous papers with the goal to introduce the method application for different cases. In that sense we practically introduce the platform for artificial neural network (ANN) application as possible solution which extend the experimental results based on different process sintering temperatures as interpolation and extrapolation approach to get some additional results designed by curvatures and diagrams with no more experiments. This is very useful from the experimental point of view because we support the measurements with additional results which practically correspond to much wider extended experimental intervals with exemptible neglected error.

Regarding previous introduction in the main subject, we make a brief cross-section of some our main previous results.

The ANN are systems inspired by neural networks of living organisms. They consist of artificial neurons-nodes that mimic basic function of biological neurons. They perform data mapping based on training process without a mathematical process model [6]. The only data necessary for mapping are input-desired output data pairs. Mapping with predefined error margin is achieved by adjusting network coefficients through the training process. Only necessary information during tranining process [7] is a network error that occurs at network output caused by incorrect network coefficients values, w_{ij}, fig. 1, [8]. On this fig. 1 neurons: *i, j* are presented by nodes while inputs are on arrows directed towards nodes and nodes outputs are on arrows directed forward. Changing coefficient values through training process leads to a decrease in the error and after training process is completed, the network mapping is satisfactory even for a new input data on network has not been trained. Training process implies changing all network coefficients starting from network output through the whole network to the input. Since this process performs from network output to network input it is called error back propagation [9]. In this way contribution of all network elements generating output error is calculated.

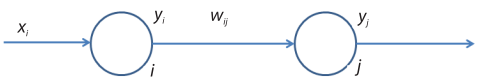

Figure 1. Nodes of a neural network

The developed technological processes and designing methods for the ceramic materials are on the way to create the full control on grains, pores and their bilayers between the constituents (grains and pores) of ceramics structures. It is very important to connect all of these intergranular phenomena including the microcapacitances by a neural network with the task to compare the results in the bulk samples measurements frame and microelectronics parameters at the micro-structure level.

The basic research was to develop the interface coating around the grains and to control the layers between two grains, as a media for electronic parameters integrations [10]. The experiments were based on nano $BaTiO_3$ powders with *Y* additives. The dielectric parameter results at the submicron level are the part of characteristic values measured at the bulk samples surfaces. The main idea is to develop the new computing methods to network electronic parameters within layers between the grains and pores to get and compare the global values at the

samples surface. The procedure of propagating an error signal through the whole network is proposed as a tool for propagating any signal of interess through the network structure that mimics ceramics structure.

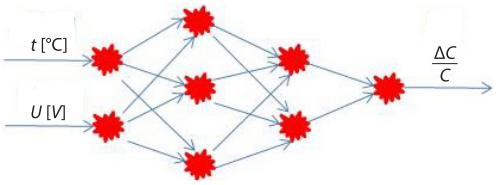

Figure 2. The principle scheme neural networks application on input parameters temperature and voltage and output parameter relative capacitance

Material structure is assumed as a multi thin layers coating around the both sides grains interconnections. Any signal measured on the material surface could be propagated throughout the structure instead of a neural network error. This idea was analysed in [11] where relative capacitance measured on a sample surface was propagated through the ceramics structure assuming that ceramic structure can be presented by a neural network, fig. 2.

Also, in addition, this structure could be expressed by a graph method as well. In this case, the grains and pores, as basic elements of material structure, can be designed by graph nodes and their mutual interconnection-lines [12]. The other aspect of this method is a common to present the ANN certain types by a graph, too. These ANN are used to present ceramic structures. The properties prognosis could be done by using neural networks. We now present one characteristic neural network as more descriptive approach for better understanding of previous explanations.

The concept of ANN and graph theory application is a part of our extended research in the field of electronic ceramic materials, sintering science, and micro-structure analysis and related phenomena especially based on fractal nature analysis and involved corrections [13-16].

These papers were the basis for further investigations on neural networks application in the sintering process analysis.

Experiment

The preparation of ceramic powder, consolidation and sintering, is a complex process to get of $BaTiO_3$-ceramics samples is presented on fig. 3. [17]. In this paper, we disposed the global scientific-technological-research algorithm with associate laboratory processes. We used high purity commercial $BaTiO_3$ Murata powder (99.9% purity, mean grain size <2 μm). For further research related to nanostructural and thermophysical properties, it was necessary to prepare the $BaTiO_3$-ceramics samples. In our research we analysed the influence of sintering temperature and time on the final $BaTiO_3$-ceramics characteristics. The $BaTiO_3$ with additive powders mixture was processed into a mill with balls and water. The organic binders were added and homogenization was about 48 hours, and the mass was transported by the membrane pump and dried, so we determined the powder granulation. The material density was tested every hour by a special vessel and after that was performed vibrating sieve. The powder particles were roughly shaped (diameters ranging 10-130 μm).

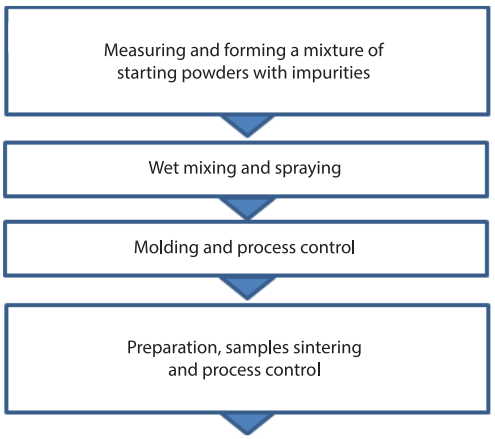

Figure 3. Scheme of the process of consolidation of $BaTiO_3$-ceramics

Along the influence of sintering temperatures, we analyzed the impact of different additives concentrations. In this research we used: CeO_2, $MnCO_3$, Bi_2O_3, Fe_2O_3. Sintering times where 2 hours, 3 hours at a temperature (1190-1370 °C) and at a pressing pressures (86-150 MPa).

Within samples testing we characterized different thermophysical and structural properties, tab.1.

Because of comparison, we applied our methods at the pure and with additives $BaTiO_3$.

Table 1. Process parameters: pressure, p, sintering temperature T_{sint}, time, τ_{sint}; pure $BaTiO_3$ and with additives

Sample type	P [MPa]	T_{sint} [°C]	t_{sint} [hour]
I-16 $BaTiO_3$-ceramics* with no additives	86	1190	2
	105	1190	2
	130	1190	2
	150	1190	2
	86	1290	2
	105	1290	2
	130	1290	2
	150	1290	2
	86	1370	2
	105	1370	2
	130	1370	2
Basic mixture $BaTiO_3$-ceramics: composition: 0.1% CeO_2 + 0.14% $MnCO_3$		1190	2
		1190	2
		1190	2
		1190	2
		1370	2
		1370	2
		1370	2
		1370	2
Basic mixture $BaTiO_3$ -ceramics: composition: 0.1% CeO_2 + 0.14% $MnCO_3$		1190	2
		1240	2
		1290	2
		1370	2
		1190	3
		1240	3
		1290	3
		1370	3

* Only for pure BaTiO3-ceramics with no additives influence of sintering time was analysed.

Apply of linear method section measuring for investigation of the influence of sintering temperature and time on grain size, l, is here presented.

Let a set: l_1, l_2,..., l_n presents discrete set of grain sizes obtained by measuring sections method. Based on relative frequencies, f_{ri}, distribution function can be empirically defined:

$$F_n^{(s)}(l)=\begin{cases}0 & l<\dfrac{C_0+C_1}{2}\\ \displaystyle\sum_{i=1}^{k} f_{ri}\,\dfrac{C_{k-1}+C_k}{2} & l\le l<\dfrac{C_k+C_{k+1}}{2},\ \ k=1,2,\ldots,s-1\\ 0 & l\ge\dfrac{C_{s-1}+C_s}{2}\end{cases} \tag{1}$$

While variation intervals $l_{\min}$, $l_{\max}$ were separated into s classes with no intersections: $[C_k, C_1)$, $[C_1, C_2)$,..., $[C_{s-1}, C_s)$. Analyzing obtained samples results it can be concluded that possible values of grain sizes interval is 0-22 μm. Classes of grain sizes are defined (class width 1.5 μm) as well as corresponding distribution functions, absolute frequencies for defined sintering temperatures and times 2 and 3 hours, respectively, tab. 2. Analyzing cumulative frequencies obtained based on appropriate absolute frequencies and appropriate functions $F_n^{(s)}(l)$, empiric function can be established:

$$F(l)=a\left[1-\mathrm{e}^{-b\left(l-l^*\right)}\right] \tag{2}$$

Table 2. Grains size as a function of a sintering temperature and sintering time

From	[μm]	2	3	2	3	2	3	2	3
0.5	1.5	149	78	58	7	45	2	2	72
1.5	3	209	179	186	104	132	36	41	51
3	4.5	60	167	104	160	101	67	69	75
4.5	6	24	52	48	94	74	82	83	89
6	7.5	7	23	34	47	37	86	59	59
7.5	9	1	7	9	17	25	52	38	48
9.0	10.5	–	3	3	13	9	36	43	40
10.5	12	–	1	6	5	12	26	30	31
12	135	–	–	1	1	6	12	25	18
13.5	15	–	–	0	2	2	18	17	18
15	16.5	–	–	0		3	10	12	7
16.5	18	–	–	1	–	3	11	10	5
18	19.5	–	–	–	–	1	4	7	4
19.5	21	–	–	–	–	–	4	7	3
21	22.5	–	–	–	–	–	–	3	3

This relation where a, b are parameters and l^* characteristic value of grain size with parameters values a, b, l^* defined in tab. 3 presents relation of grain size according to temperature and sintering time. On fig. 4 is a presentation of results obtained from tab. 2 applying eq. (1).

It can be noticed that on lower sintering temperatures typical grain sizes are within narrow interval so the grain size value of 10 μm presents critical value because there is no grain size value greater then it (flattened part of the curve). On the sintering temperature of 1290 °C the result is quite different since the range of values is much wider (smaller slope of the curve). On the sintering temperature of 1370 °C this characteristic is more expressed since flattening

appears for values greater then 25 μm so this curve slope is significantly lower then all other curves. Similar relation is presented on fig. 5 where it could be noticed that longer sintering time enables wider interval of possible values of grain sizes on lower temperatures as well. This phenomena can be explained by influence of sintering time and material density increase as a consequence of sintering temperature increase.

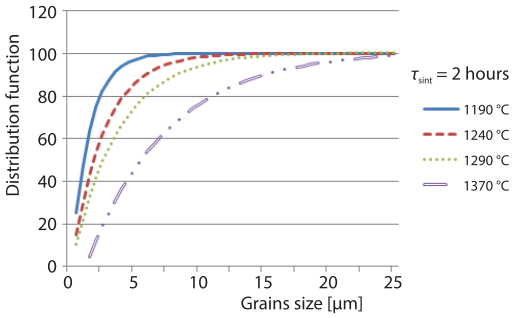

Figure 4. Distribution function for different sintering temperatures, τ_{sint} = 2 hours

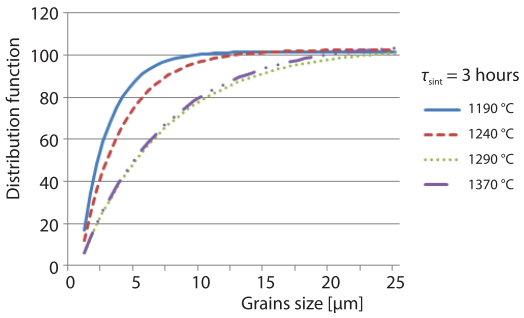

Figure 5. Distribution function for different sintering temperatures, τ_{sint} = 3 hours

By different micro-structural and qualitative metallography methods we have got the cumulative function parameters values, tab. 3.

Table 3. Cumulative function parameters values

T [°C]	t [hour]	a	b [μm^{-1}]	l^* [μm]
1190	2 3	100.2 101.7	0.72 0.46	0.1 0.6
1240	2 3	100.2 102.5	0.40 0.31	0.1 0.6
1290	2 3	100.5 103.7	0.28 0.15	0.1 0.6
1370	2 3	101.2 106.1	0.16 0.15	1.2 0.6

It is evident that number of grains per area unit decreases namely, grains size increases as sintering temperature increases from 1190 °C, 1240 °C, 1290 °C up to 1370 °C. The grains growth is noticed at the sintering temperature of 1290 °C.

The effects of sintering temperature increase have the influence on sintering process and by that on ceramic material micro-structural constituents. In the case of same sintering time, as well as material density (pressing pressure) sintering temperature increase positively effects on $BaTiO_3$ ceramics consolidation.

One can notice that extremely characteristic *breaking* character of sintering temperature 1240 °C relative to other sintering temperatures. The other methods (SEM and optical microscopy) confirm the influence of sintering temperature of 1240 °C as a key factor in sintering process namely $BaTiO_3$-ceramic material total consolidation process.

Results and discussion – advantages of the ANN based approach

In the previous section the significance of sintering temperature to ceramics consolidation has been observed. Conclusions were drawn based on the experiments performed on

different certain sintering temperatures (1190 °C, 1240 °C, 1290 °C, and 1370 °C) using distribution function. It would be of interest to define a distribution function for an arbitrary sintering temperature. This can be performed by extrapolating and interpolating experimental measurements [18,19]. One of possible procedure is the use of neural networks [20]. From now on, we present the application of a back-propagation neural network that solves this problem.

Back propagation neural network can potentially perform arbitrarily input-output data mapping. Desired input-output mapping is known in advance. This neural network possesses numerous adjustable parameters – weight coefficients (weights) which have to be set on appropriate values to provide accurate mapping. Since weights starting values are set at random input-output mapping is incorrect. Adjusting the weight coefficients on their appropriate values to provide an accurate mapping is called the training process. This implies the introduction of input data to neural network which generates neural network output. Output error occurs as a result of incorrect network weight values. All weights are changed backward through the network, starting with output layer neurons, continuing with hidden layer neurons to the input layer neurons. This change provides network error reduction. This procedure is repeated numerous times for all input-output data. Network is trained when input-output data set is mapped under predefined error. As a result of the training process, the neural network is able to map the new input data with satisfactory accuracy.

In general, main idea in this paper is to develop the approach which consists in combining neural networks and interpolation method, to get the parameters defined in tab. 3. a, b, l^* from distribution function data. After getting these parameters and completing the distribution function, we can easily produce another, fourth curve which practically confirms this original combined methods application as a successful with very low error. This way we opened new practical frontiers for designing and predicting experimental tasks and desired results. All this physical-mathematical experimental-theoretical combined method makes the procedure much more efficient for scientists and their experiments in the science of sintering.

Neural network extrapolation

Network is trained to present distribution function eq. (1), for an arbitrary sintering temperature based on the set of four experimental results, for sintering time of 2 hours, fig 4. The curve parameters are shown on tab. 3. Input signals are 146 points on each curve representing grain size and appropriate distribution function values: l, F, (l). These 2-D vectors are presented to neural network as an input. Neural network had one hidden layer with 10 neurons and 3-D output layer, fig. 6.

Desired outputs of the network are constants during the training process for all the input data of a curve, the desired outputs are unchanging. For all 146 2-D input vectors of a curve for 1190 °C, tab. 3, first row, desired output vector should be x = (100.2; 0.72; 0.1) while for other input temperatures: 1240 °C, 1290 °C, and 1370 °C output vectors are from tab. 3 defined, respectively.

For the sake of testing neural network capability to predict curve parameters training will be performed on three experimental curves while fourth will be a test input. Network was

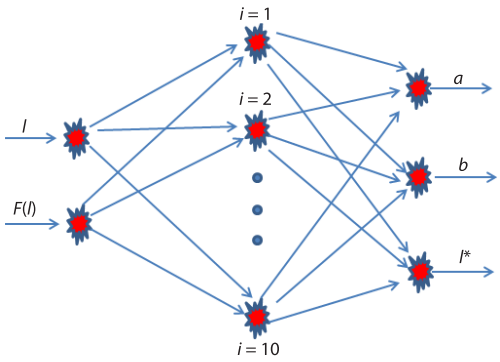

Figure 6. Neural network structure

trained on data presenting distribution function for 1240 °C, 1290 °C, and 1370 °C while curve for 1190 °C was used for testing. After training process is finished testing was performed by choosing a point from a test curve (for a 1190 °C it was l, $F(l) = 5; 97.3$). Distribution function parameters a, b, l^* were obtained a with mean squared error: $MSE = 1.55 \cdot 10^{-1}$. Further on, network was trained on data presenting 1190 °C, 1240 °C, and 1290 °C while a point from a curve for 1370 °C was used for testing (l, $F(l) = 8; 67.1$). Mean squared error calculating parameters a, b, l^* of a curve for 1370 °C was in this case: $MSE = 7.02 \cdot 10^{-1}$.

Neural network interpolation

In this case, network was trained on curves representing sintering temperatures: 1190 °C, 1240 °C, and 1370 °C while test curve is 1290 °C. After training process is finished testing was performed by choosing a point from a test curve (for a 1290 °C it was l, $F(l) = 5; 75.01$). For that point parameters a, b, l^* were calculated with mean squared error $MSE = 3.3 \cdot 10^{-2}$.

Further on network was trained on data presenting 1190 °C, 1290 °C, and 1370 °C while curve for 1240 °C was used for testing. In this case parameters a, b, l^* were calculated for l, $F(l) = 3; 68.79$ with mean squared error $MSE = 4.7 \cdot 10^{-2}$.

In comparison with a combination of neural network and interpolation, where the errors are below 10^{-2}, by extrapolation we have errors on the level 10^{-1}. This is also a still acceptable errors frame that we can successfully apply neural networks as we already done by interpolation.

Outlook

In future research, we plan to develop application neural networks for other sintering consolidation parameters, not only for sintering temperature, but also for sintering time, and pressure. We will consider the creation of some software application, which can contribute on direction of these methods.

Conclusion

In this paper we applied the neural networks method with interpolation and extrapolation methods within sintering temperature interval 1190-1370 °C. In this case we used 2 hours sintering time at the main sintering zone. We got results on the way for experimental-theoretical combined neural networks and interpolation-extrapolation methods application, which confirm successful idea. It has been established that greater accuracy is achieved applying neural networks as interpolation than extrapolationol. Namely, extrapolation of distribution function is achieved with MSE order of magnitude – 1 while interpolation shows that MSE is order of magnitude – 2. Such quantitative difference between results of extrapolation and interpolation was expected since extrapolation is quantitively a subject of greater uncertainty. Both results achieve acceptable errors and present basis for further research. In sintering science, the existing practice means that we must always do many experiments, which sometimes means a waste of time and does not provide high level of experimental efficiency. Based on the results in this paper, we have opened new frontiers in the prognosis of experimental parameters and desired results from the experiment, which is more efficient and faster way on how to plan and design the results. Also this is very helpful in the sense of getting the results and curves in experimental temperature space-interval between the previous known results, what could be helpful in experimental development, to do or to avoid some experiments in between of existing interval points what is very important in experimental programming. The most advanced trend in sintering

process development is decreasing the sintering temperature in direction of energy and production time and efficiency. A novelty of our ANN method application in the sintering process is that we can shortcut the experimental and producing procedure due to the projected temperature that we can apply with very neglectable errors. This is a truly advanced and original innovation of the sintering process application.

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