THE OPTIMALIZATION OF THE PROPERTIES OF THE MULTICOMPONENT PZT TYPE CERAMICS BY THE CHOICE OF APPROPRIATE TECHNOLOGICAL CONDITIONS

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1. INTRODUCTION

The PZT-type materials are commonly used in engineering. They are used as transducers between mechanical and electrical energy, such as phonograph pickups, air transducers, underwater sound and ultrasonic generators, delay-line transducers, wave filters, piezoelectric micromotors, microrobots, actuators, etc. [1,2]. The achievements of the technology and researches of the new ceramic ferroelectric materials lead to progress in the recently piezoelectronic. The exploration and study of the piezoelectric effect in PZT ceramics makes huge possibility of building piezoceramic electronic elements. The increasing possibilities of applications of these materials are connected with both the selection and improvement of the structure and microstructure (decreasing porosity, increasing density, decreasing grain dimensions) by means of choice of the suitable technological conditions [3].

The multicomponent solid state solutions have better parameters than pure PZT solutions because:

- give possibility of influence on the physical properties (selection of the chemical composition);
- make better synthesis conditions (increasing of the intensification of diffusion processes);
- give possibility of choice material with the required set of parameters.

The most common widespread ceramic obtain method is conventional ceramic sintering method. Ceramic materials are obtained from simple oxides (milling, synthesis, sintering) in this method. The PZT type ceramics synthesis most frequently are made by the traditional method reaction in solid state in about 1223 K temperature. The most important part of preparation powder to synthesis is mixing components. It’s caused that chemical composition and microstructure are very homogeneous. At the beginning synthesis have place at the nearest grain boundaries of the mixture components. The reactions in the solid state mainly have place on the surface of the material therefore purity of the initial components and the level of mixing has significant influence for synthesis process. During the growth of the grain size of the new phase rapidity of the reaction is decreasing. It's caused by decreasing of the rapidity of mass diffusion. Intensify of the synthesis process could be done by the interruption of this process and repeating of milling, mixing and sintering once again. Increasing of rapidity of the diffusion process could be done in two ways: growth of surface of the grain (milling) or modification of the condition of surface (mechanical process). The chemical synthesis method (sol-gel) is used for decreasing of the way of the diffusion of molecule [4,5].

All of the technological parts process (milling, mixing, synthesis, sintering, mechanical process, depositing electrodes and polarization) decided to ceramics properties [6-9]. In the multicomponent ceramic materials process of the origin of the final material have happen in many parts. In the reason of that two part synthesis came to obtain materials with better properties.

In this work have shown the result of the investigation of the dielectric and piezoelectric properties multicomponent PZT type ceramics obtained in two ways: synthesis one and multi parts.

2. MATERIAL AND INVESTIGATION METHOD

The ceramics with the following chemical composition $0{,}55PbTiO_3$-$0{,}43PbZrO_3$-$0{,}02[Pb(Cd_{0,5}W_{0,5})O_3]$ has been investigated. Two series of sample (A and B) were obtained by the conventional ceramic sintering method. The samples were obtained from oxides PbO, ZrO_2, TiO_2, CdO, WO_3 milled through 15 hours. In the case of series A synthesis were in 1223 K temperature by 3 hours, however series B synthesis were in 1223 K by 6 hours (two parts, each 3 hours long). Both series were sintered in 1423 K temperature by 6 hours.

A very important question in the technology of ferroelectric materials is to provide consistence in the stoichiometry between product obtained and chemical composition described by a molecular formula of the compounds. It's of great importance in the case of ceramic ferroelectrics containing lead [10]. Therefore sintering were in presence of the casting powder with the same chemical composition as investigated material with 5% excess of PbO.

The samples were ground and polished. Electrodes on their surface were deposited by the silver paste burning method. All samples were subjected to polarization by the low temperature method (T_{pol}=423 K, E_p=30 kV/cm, t_{pol}=30 min). Ceramic samples in the shape of discs, to measurements piezoelectric properties (resonance-antiresonance method) and dielectric properties (capacity bridge of the BM 595 Tesla type) were used.

3. RESULTS AND DISCUSSION

Measurements of dielectric permittivity ε and tangent of dielectric loss of angle tgδ were obtained for 55% mol $PbTiO_3$ ceramic materials to show how conditions of synthesis have an influence on dielectric properties.

All curves of dielectric permittivity $\varepsilon_{33}^{T}/\varepsilon_0(T)$ obtained with various frequencies for ceramics with different times of synthesis A (3 hours) have clear maximum connected with phase transition which take place at the temperature 586 K.

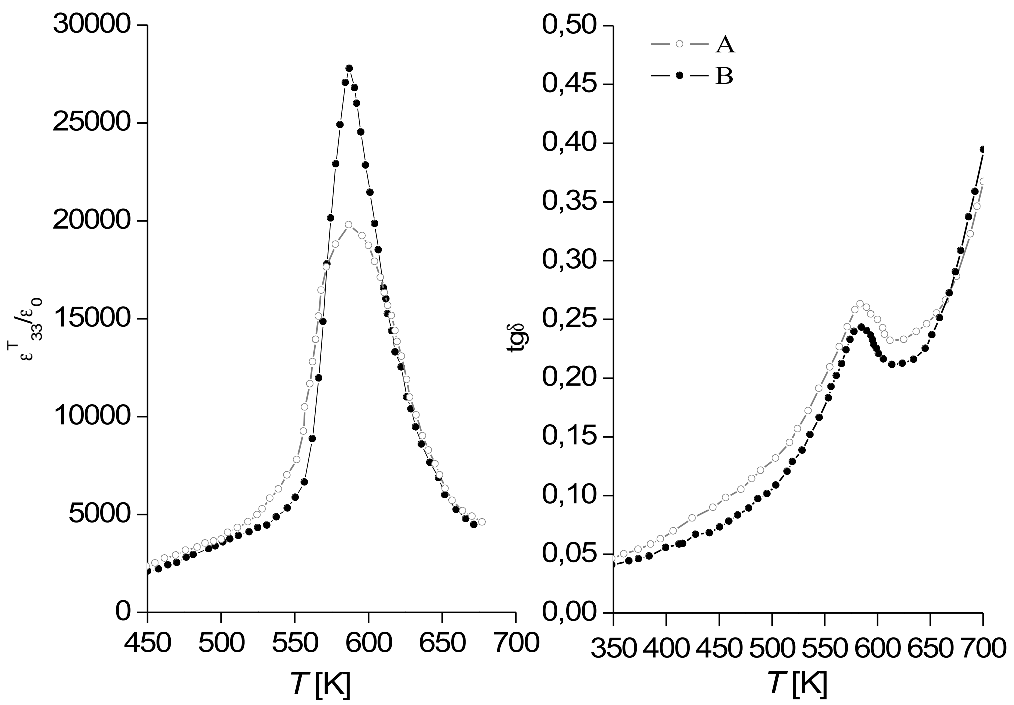

Fig. 1. The temperature dependencies of dielectric permittivity ε and tangent of dielectric loss of angle tgδ for the samples with different synthesis time A (3h) and B (6h).

The lack of temperature displacement of maximum of dielectric permittivity with changing frequencies proves that transition between ferroelectric and paraelectric phase is the 1st type transformation. The same situation was observed for ceramic samples B (6 hours of synthesis).

The samples with synthesis time A (3h) have lower values of the dielectric permittivity and much higher values of the tangent of dielectric loss of angle both in the room temperature and in the Curie temperature (fig 2.). We also could observed that transition takes place in wider range of temperatures and the peak of dielectric permittivity become wider. It's caused by not completely reaction of components (oxides) and it is a result of short time synthesis, too.

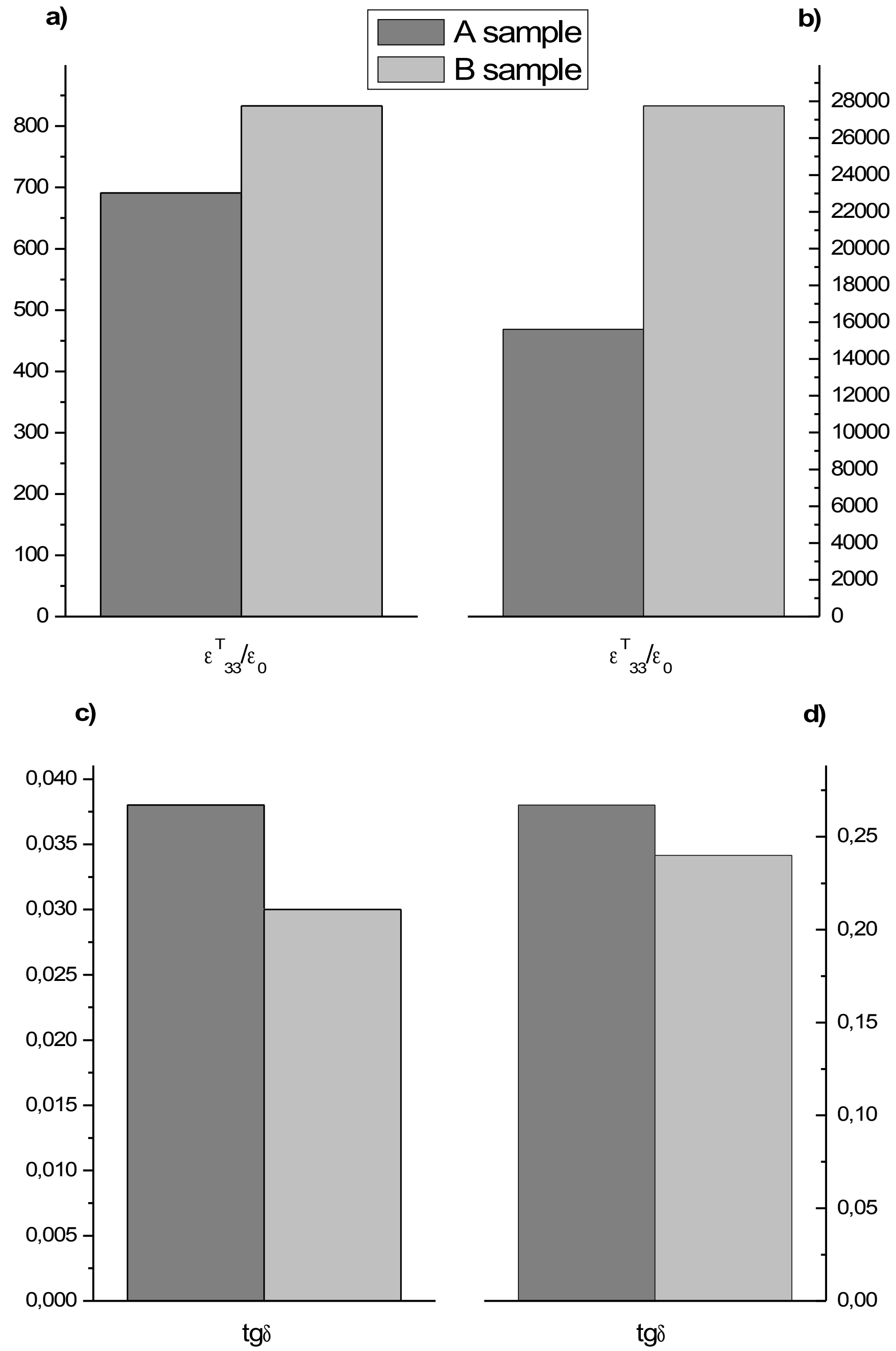

Fig. 2. The influence of synthesis time on dielectric properties: a), c) room temperature 294K, b), d) in the Curie temperature.

The SEM images from the crack of the samples A (fig. 3a) and B (fig. 3b) have been made in Laboratory of Field Emission Scanning Electron Microscopy and Microanalysis at the Institute of Geological Science of the Jagiellonian University.

a) b)

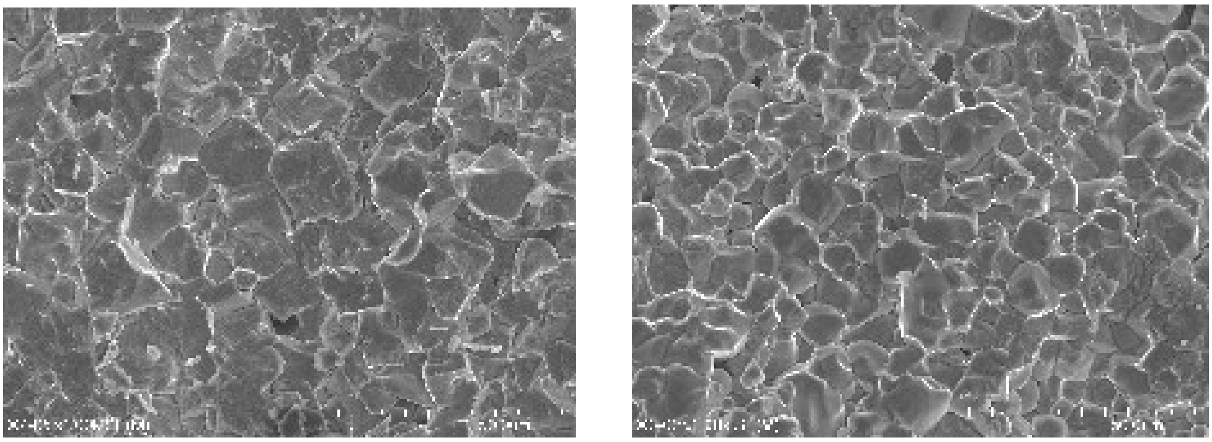

Fig. 3. The SEM images from the crack of the samples: a) A (3h long synthesis), b) B (6h long synthesis).

The SEM images show not enough developed grains. As a result of that crack took place along the grains not as usually happens along the grains boundaries (fig. 3).

Table I. Piezoelectric and dielectric parameters of the samples.

Samples obtained by conventional ceramic sintering method		
Parameter type	**Sample A**	**Sample B**
density ρ [kg/m^3]	6170	7243
piezoelectric modulus $d_{31} \times 10^{12}$ [C/N]	56,76	71,62
electromechanical coupling coefficient k_p	0,3432	0,4651
acoustic velocity V_R [m/s]	2107	2512
elastic susceptibility $S^E_{11} \times 10^{12}$ [m^2/N]	12,97	12,93
dielectric permittivity $\varepsilon^T_{33}/\varepsilon_0$	691	833
tangent of dielectric loss of angle tgδ	0,038	0,03

In the table I piezoelectric and dielectric parameters are presented. Higher values of density, piezoelectric modulus and electromechanical coupling coefficient were observed in samples B which had longer synthesis time (6h). It’s caused by smaller grains size obtained by the way of double synthesis. The acoustic velocity in sample A has lower value than acoustic velocity in sample B because sample A has a lower density and it’s common known that acoustic velocity is inversely proportional connected with density.

The decrease of values of elastic susceptibility and tangent of dielectric loss of angle and increase of dielectric permittivity in sample B are another proofs that longer sintering have immense influence to properties of piezoelectric ceramics.

4. CONCLUSIONS

1. An increase of the synthesis time in the PZT type ceramics has an influence on grains size and development of grains.
2. Longer synthesis time lead to obtain materials with better dielectric and piezoelectric properties such as permittivity ε, tangent of dielectric loss of angle tgδ, electromechanical coupling coefficient k_p and piezoelectric modulus d_{31}.
3. Changes in the time of synthesis in multicomponent ceramics of the PZT type lead to obtain materials for different range of technical applications.

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