THE FERROELECTRIC PLZT TYPE CERAMICS AS A MATERIAL FOR TRANSDUCERS

M. CZERWIEC, R. ZACHARIASZ, J. ILCZUK
University of Silesia, Faculty of Computers Science and Materials Science,
Department of Material Science
3 Żeromskiego St, 41-200 Sosnowiec, POLAND, marek.czerwiec@orange.pl

(*received.....; accepted.....*)

Modification of the PZT system by the addition La^{3+} ions has marked beneficial effect on several the basic parameters, such as squerness of the hysteresis loop, decreased coercive filed, increased dielectric constant, maximum coupling coefficients, increased mechanical compliance, and enhanced optical transparency.
The mechanical and electrical properties in lanthanum modified lead zirconate-titanate ceramics of 5/50/50 and 10/50/50 were studied by electric permittivity ε and tangent of dielectric loss of angle tgδ measurements. The temperature dependences of ε=f(T) and tgδ=f(T) were determinate in temperature range from 300 K to 730 K. The values of T_C obtained during ε and tgδ measurements were respectively: 560 K for 5/50/50 and 419 K for 10/50/50.

Key words: PLZT, electric permittivity, tangent of dielectric loss of angle.

1. Introduction

$Pb(Zr, Ti)O_3$ (PZT) crystallizes with the ABO_3 type structure in which the A-site is occupied by Pb^{2+} ions; Zr^{4+} and Ti^{4+} are accommodated on the B-site. The influence of various substitutions in the A and B-site of PZT unit cell has been studied by numerous investigators. The PLZT formula ($Pb_{1-x}La_x(Zr_yTi_{1-y})_{1-x/4}O_3$) assumes that La^{3+} substitutes for Pb^{2+} in the A-site and the B-site vacancies are created for electrical balance. The composition of PLZT is routinely represented by the notation *x*/(1-*y*)/*y*, which denotes the amount of La/Zr/Ti, given in mole fractions or mole per cent. The PLZT type ceramic may be used as a materials for device applications such as non-volatile memories, transducers, modulators, etc. [1-3].

2. Experiment

The aim of this work was to obtain solid solution of the PLZT from ferroelectric phase with constant ratio Zr/Ti=50/50 and variable concentration of La^{3+} ions:

- **$Pb_{0,95}La_{0,05}(Zr_{50}Ti_{50})_{0,9875}O_3$ - PLZT 5/50/50**

- **$Pb_{0,95}La_{0,10}(Zr_{50}Ti_{50})_{0,975}O_3$ - PLZT 10/50/50**

and investigate electromechanical properties of obtained ceramics.

Ceramic samples were obtained as a reaction in solid state from simple oxides: PbO, ZrO_2, TiO_2, La_2O_3 by conventional ceramic sintering (CCS) method. Ceramic powders were mixed and milled through 20h and next formed in cylindrical tablets of diameter 10 mm. After this tablets were synthesized at the temperature T_S = 1123K through t_s = 6h. Then polycrystalline samples were crumbled and mixed to obtain more homogenous structure. The samples in a shape of rectangular bars form were received. Next, all samples were ground and polished to the dimensions (30x10x0,9) mm^3 and then electrodes were deposited on their surface by the silver paste burning method. The samples in a shape of discs (10x1 mm^2) were obtained too. The obtained samples were subjected to polarization using the low temperature method at 423 K for 30 min.; the intensity of the polarization field was Ep=30 kV/cm [4]. The measurement of dielectric permittivity ε and tangent of dielectric loss of angle $tg\delta$ were obtained by the capacity bridge BM 507/538 Tesla type with frequency 1 kHz and temperature range between 300 K and 730 K.

3. Result and discussion

The measurements of the temperature dependences of $\varepsilon(T)$ and $tg\delta(T)$ were obtained as a aim of detailed analysis of the changes in the area of phase transition. The results of investigation are shown at the figure 1 & 2. The nature of the temperature dependences of $tg\delta(T)$ in the range of temperatures below phase transition is connected with dissipation of energy to polarization of the domains. But above the phase transition temperature (T_C) losses of energy are related with electric conductivity. For both chemical composition of PLZT type ceramics the temperature dependences of $\varepsilon(T)$ has a relaxor character with diffuse phase transition between ferroelectric and paraelectric phase.

The measurements of parameters characterizing piezoelectric properties of the ferroelectric ceramic were made by the resonance-antiresonance method [5]. Using the resonance-antiresonance method the electromechanical coupling coefficient (k_p) for samples in the shape of discs can be calculated by the following formula:

$$k_p = \sqrt{\frac{n^2 - 1 + \nu^2}{2(1+\nu)}\left(1 - \frac{f_r^2}{f_a^2}\right)}, \tag{1}$$

where: f_a - antiresonance frequency [kHz],
f_r - resonance frequency [kHz],
ν - Poisson's ratio,
n - the lowest positive root of molecular equation.

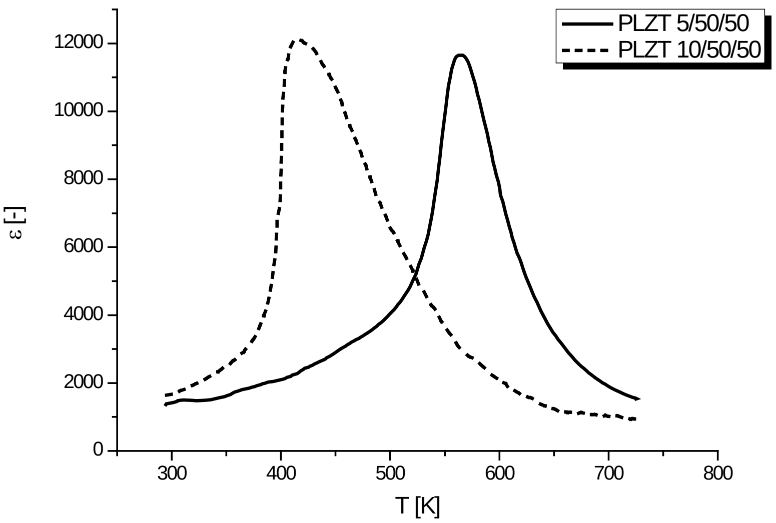

Fig. 1. The temperature dependences of ε(T) and tgδ(T) for PLZT tested samples.

The k_p coefficient characterizes a part of electric energy transformed into mechanical energy while applying an external electric field. It can also characterizes a part of mechanical energy transformed into the electric energy, in a case when the mechanical stress is applied to the samples. The k_p value is always lower than unit, because during transformation of one type of energy into other a part of energy is dispersed. The values of k_p coefficient as well as for PLZT 5/50/50 and PLZT 10/50/50 are almost the same and respectively are 0,33 and 0,32.

In order to determine the piezoelectric modulus d_{31} the resonance frequency f_r of radial vibrations was found and then the following relationship was used:

$$d_{31} = \frac{0{,}188 \cdot k_p}{\frac{d}{2} f_r} \sqrt{\frac{\varepsilon}{\rho}}, \qquad (2)$$

where: k_p - the electromechanical coupling coefficient,

ε - the tensor's component of electric permittivity,

ρ - density of the sample,

d - diameter of the sample.

To calculate the values of acoustic velocity V_R the following formula was used:

$$V_R = \frac{f_R \cdot 2\pi r}{n}, \tag{3}$$

where: r - the radius of the smaple,
n - the lowest positive root of molecular equation.
$f_R = f_a - f_r$.

The values of elastic susceptibility were calculated from the relationship:

$$S_{11}^E = \frac{2 \cdot (d_{31})^2}{(k_p)^2 \cdot (1-\nu) \cdot \varepsilon \cdot \varepsilon_0}, \tag{4}$$

where: ε_0 - permittivity in the vacuum.

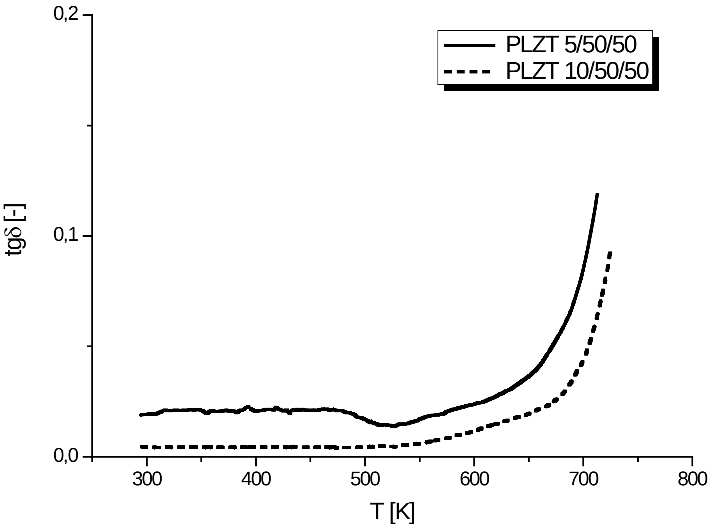

Fig. 2. The temperature dependences of tgδ(T) for PLZT tested samples.

Table 1. Basic parameters measured at 293K.

Parameter	Value	
% mol La	5	10
electromechanical coupling coefficient k_p [-]	0,33	0,32
cross electromechanical coupling coefficient k_{31} [-]	0,17	0,18
piezoelectric modulus $d_{31} \cdot 10^{11}$ [C/N]	2,24	6,36
acoustic velocity V_R [m/s]	2446	2030
elastic susceptibility $S_{11}^E \cdot 10^{11}$ [m^2/N]	1,61	1,32
elastic susceptibility $S_{12}^E \cdot 10^{12}$ [m^2/N]	-7,53	-5,06
elastic modulus $C_{11}^E \cdot 10^{-10}$ [N/m^2]	6,17	7,56
density ρ [kg/m^3]	7542	7315
Poisson's ratio υ [-]	0,46	0,38
modulus g_{31} [Vm/N]	0,021	0,0068
resonance frequency f_r [kHz]	229	266,2
antiresonance frequency f_a [kHz]	239,1	278,1
1st Overton's frequency f_r [kHz]	620	689,4

4. Conclusions

Obtained PLZT type ceramic samples are characterized by low values of tangent of dielectric loss of angle tgδ=1÷1,5% and high values of electric permittivity ε=11000÷12000 (at room temperature). The measurement of the temperature dependences of ε let to obtained the temperature of phase transition (T_C). The value of T_C is decreasing with increasing of La content. Lanthanum has also significant influence for level of diffuse phase transition.

References

[1] G. H. Haertling, J. Am. Ceram. Soc. **82** (4) (1999) 797.

[2] C. L. Hulang, B. H. Chen, Wu L., Solid State Comm. **130** (2004) 19.

[3]. E. M Bourim, H. Tanaka, M. Gabbay, G. Fantozzi, B. L. Cheng, J.Appl. Phys. **91** (2002) 6662.

[4] R. Zachariasz, B. Bruś, J. Bluszcz, Molecular and Quantum Acoustics **24**, (2003) 249.

[5] B. Bruś, J. Ilczuk, Molecular and Quantum Acoustics **23**, (2002) 89.