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The temperature dependences of electromechanical properties of PLZT ceramics

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Abstract. The mechanical and electrical properties in lanthanum modified lead zirconate-titanate ceramics of 5/50/50 and 10/50/50 were studied by mechanical loss Q^{-1} , Young's modulus E , electric permittivity ϵ and tangent of dielectric loss of angle $\text{tg}\delta$ measurements.

The internal friction Q^{-1} and Young modulus E measured from 290 K to 600 K shows that Curie temperature T_C is located at 574 K and 435 K (1st cycle of heating) respectively for ceramic samples 5/50/50 and 10/50/50. The movement of T_C in second cycle of heating to lower temperature (561 K for 5/50/50 and 420 K for 10/50/50) has been observed. Together with Q^{-1} and E measurements, temperature dependences of $\epsilon=f(T)$ and $\text{tg}\delta=f(T)$ were determinate in temperature range from 300 K to 730 K. The values of T_C obtained during ϵ and $\text{tg}\delta$ measurements were respectively: 560 K for 5/50/50 and 419 K for 10/50/50. These temperatures are almost the as temperatures obtained by internal friction Q^{-1} measurements in second cycle of heating. In ceramic sample 10/50/50 the additional maximum on internal friction Q^{-1} curve at the temperature 316 K was observed.

1. INTRODUCTION

$\text{Pb}(\text{Zr}, \text{Ti})\text{O}_3$ (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 in order to modify the properties of the materials for device applications such as non-volatile memories, transducers, modulators, etc. 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 field, increased dielectric constant, maximum coupling coefficients, increased mechanical compliance, and enhanced optical transparency [1-4].

2. EXPERIMENT

The material tested were solid solution of the PLZT from ferroelectric phase with constant ratio $\text{Zr}/\text{Ti}=50/50$ and variable concentration of La^{3+} ions : $\text{Pb}_{0,95}\text{La}_{0,05}(\text{Zr}_{50}\text{Ti}_{50})_{0,9875}\text{O}_3$ - PLZT 5/50/50 and $\text{Pb}_{0,95}\text{La}_{0,10}(\text{Zr}_{50}\text{Ti}_{50})_{0,975}\text{O}_3$ - PLZT 10/50/50.

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 $T_s = 1123\text{K}$ through $t_s = 6\text{h}$. Then polycrystalline samples were crumbled and mixed to obtained more homogenous structure. The range of sintering temperatures ($1223\text{ K} \div 1523\text{ K}$) and sintering times ($6\text{h} \div 12\text{h}$) were dependable from chemical composition. The electrodes were deposited on the surface of PLZT samples by the silver paste burning method. The obtained samples were subjected to polarization using the low temperature method ($T_p = 423\text{ K}$, $E_p = 30\text{ kV/cm}$, $t_p = 30\text{ min}$). The measurement of dielectric permittivity ϵ and tangent of dielectric loss of angle $\text{tg}\delta$ were obtained by the capacity bridge BM 507/538 Tesla type with frequency 1 kHz and temperature range between 300 K and 730 K. Temperatures dependences $Q^{-1}(T)$ and $E(T)$ were performed by automatic resonance spectrometer RAK-3 type controlled by computer.

3. RESULTS AND DISCUSSION

On the fig. 1 temperature dependences of $Q^{-1}(T)$ and $E(T)$ are presented. For the composition 5/50/50 the measurements were made during two cycle of heating with different resonance frequency (f_r) of sample vibration. In the room temperature the values of f_r were respectively: 838 Hz for 1st cycle of heating and 800 Hz for 2nd cycle.

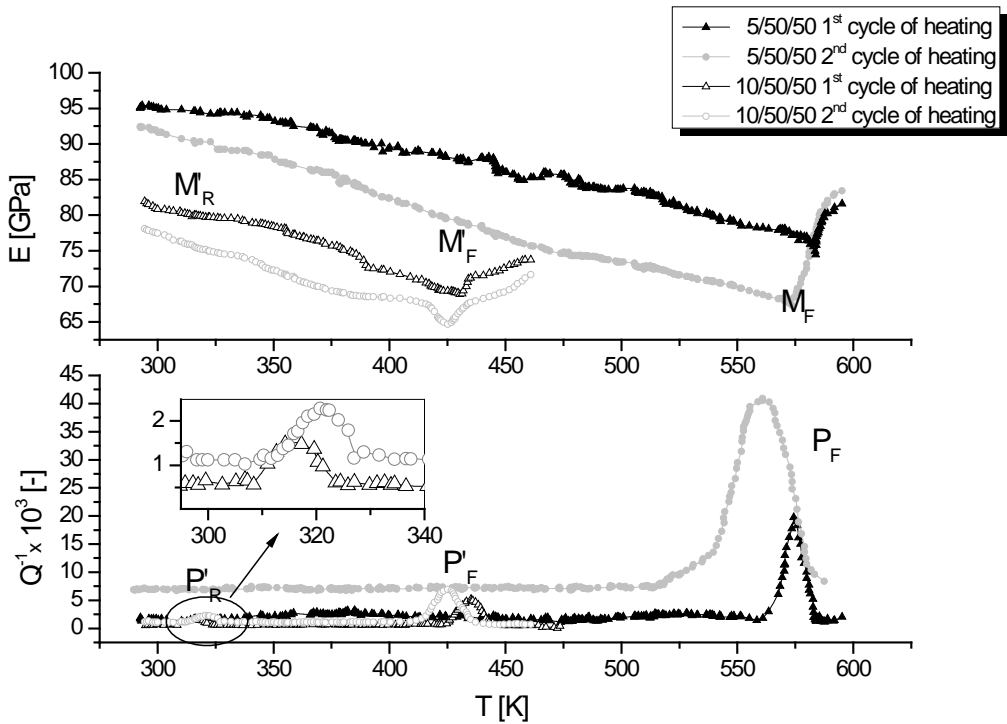


Figure 1. The temperature dependences of $Q^{-1}(T)$ and $E(T)$, for PLZT tested samples.

Similarly for 10/50/50 composition, f_r were 900 Hz and 866 Hz. All measurements were conducted with 3K/min ratio of heating. Both for samples 5/50/50 and 10/50/50 after 1st cycle of heating, clearly maxima P_F i P'_F on the curves $Q^{-1}(T)$ were observed. The maxima correlated with minima M_F i M'_F observed on $E(T)$ curves. Those changes were observed in temperatures: $T_F = 574$ K i $T_F = 435$ K. After deep analysis of phase diagram of PLZT system was proved that the temperatures are responsible for phase transition ferroelectric \leftrightarrow paraelectric (Curie temperature T_C) [2]. In the 2nd cycle of heating was observed clearly increase of height of peaks P_F i P'_F and their little movement to lower temperatures: $T_F = 561$ K for PLZT 5/50/50 and $T_F = 420$ K for PLZT 10/50/50. In the references of the earlier works authors proved that behavior like this of the maxim $Q^{-1}(T)$ in the areas of phase transitions is mainly connected with phenomena described by the Delorme and Gobin model [5,6]. Additionally for composition PLZT 10/50/50 at curve $Q^{-1}(T)$ maximum P'_R was observed in temperatures 316 K and 322 K respectively for 1st and 2nd cycle of heating. The way of behavior of this maximum and parameters calculated by the Arrhenius' law: energy of activation $H = 2,33$ eV and preexponential $\tau_0 = 5 \cdot 10^{-29}$ s⁻¹ proved that origin of the maximum is connected with more complicated processes and it will be subject of future researches.

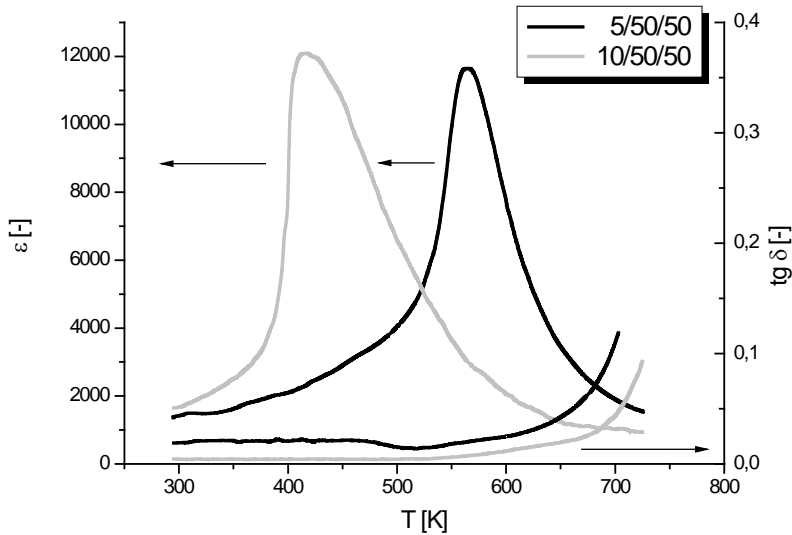


Figure 2. Zależności $\varepsilon(T)$ and $\text{tg}\delta(T)$.

The measurements of the temperature dependences of $\varepsilon(T)$ and $\text{tg}\delta(T)$ 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 2. The nature of the temperature dependences of $\text{tg}\delta(T)$ 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 linear and quadratic Curie-Weiss laws were used to analysis of the dependences of $1/\varepsilon(T)$. At the paraelectric phase above temperature T_1 (PLZT 5/50/50) i T'_1 (PLZT 10/50/50), the linear Curie-Weiss law was used [7]:

$$\varepsilon = \frac{C_{CW}^+}{T - T_m} \quad (1)$$

where: C_{CW}^+ – Curie-Weiss constant,
 In the ferroelectric phase Curie-Weiss law:

$$\epsilon = \frac{C_{CW}^-}{T_m - T} \tag{2}$$

where: C_{CW}^- – Curie-Weiss constant ,

The values of the Curie-Weiss constants as well for paraelectric and ferroelectric phase are presented in table 1. The range of temperatures T_1 - T_2 is responsible for relaxor diffuse phase transition. In the range of temperatures between T_m and T_1 experimental curves of dependences $\epsilon(T)$ are described by quadratic Curie-Weiss law [8]:

$$\frac{1}{\epsilon^*} = \frac{1}{\epsilon} - \frac{1}{\epsilon_m} = K(T - T_m)^\gamma \tag{3}$$

where: ϵ_m electric permittivity at the temperature of phase transition T_m .

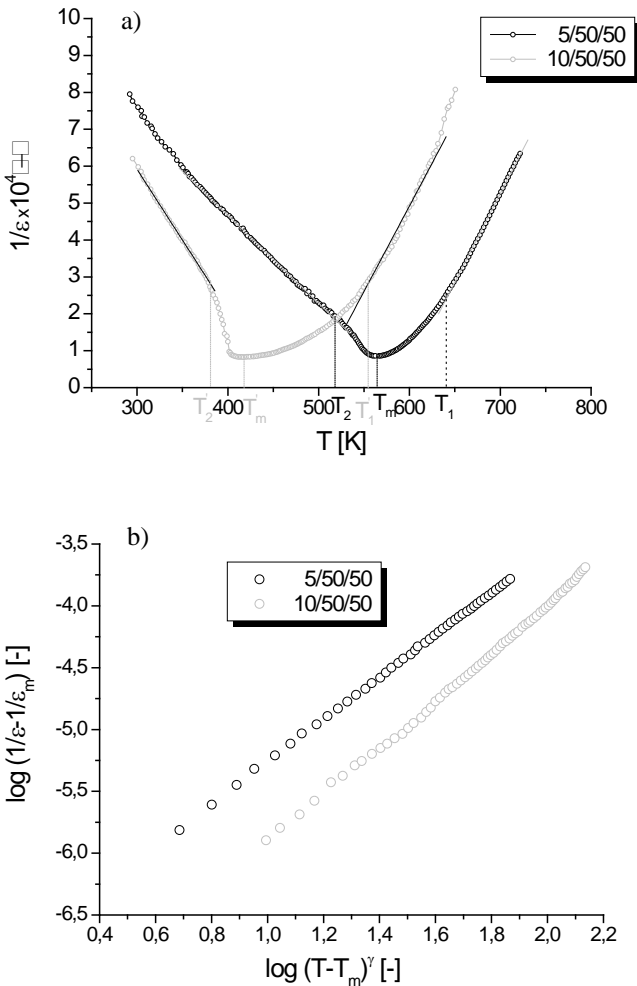


Figure 3. The dependences $1/\epsilon(T)$ fig 3a and $\log(1/\epsilon - 1/\epsilon_m)=f(\log(T - T_m)^\gamma)$ fig 3b.

4. CONCLUSIONS

Obtained PLZT type ceramic samples are characterized by low values of tangent of dielectric loss of angle $\text{tg}\delta=1\div1,5\%$ and high values of electric permittivity $\epsilon=11000\div12000$ (at room temperature). The measurement of the temperature dependences of Q^{-1} and E 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.

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