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Polarization and electrophysical parameters of piezoceramic materials investigation

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Abstract. The effect of temperature and electric field strength on the polarization and dielectric relaxation of the $Ba_{0.85}Ca_{0.15}Zr_{0.1}Ti_{0.9}O_3$ piezoceramic material has been studied. Experimental studies were carried out using the automated information-measuring system developed by the authors. It is based on the modified Sawyer–Tower circuits, which make it possible to measure the hysteresis loops of the polarization dependences on the electric field strength in the ferroelectric phase at different temperatures.

Keywords: temperature, polarization, piezoceramics

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Исследование поляризации и электрофизических параметров пьезокерамических материалов

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Аннотация. Исследовано влияние температуры и напряженности электрического поля на поляризованность и диэлектрическую релаксацию пьезокерамического материала $Ba_{0,85}Ca_{0,15}Zr_{0,1}Ti_{0,9}O_3$. Экспериментальные исследования выполнены с помощью разработанной авторами автоматизированной информационно-измерительной системы. В ее основе –модифицированные схемы Сойера – Тауэра, которые позволяют измерять петли гистерезиса зависимостей поляризованности от напряженности электрического поля в сегнетоэлектрической фазе при разных температурах.

Ключевые слова: температура, поляризация, пьезокерамика

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Introduction

Materials based on barium titanate ($BaTiO_3$) are promising piezomaterials. It has been found that the dielectric and piezoelectric properties of $BaTiO_3$ can be improved by adding elements such as Ca, Sr, Mg or Zr. It is stated in [1] that the BCZT ceramics having the composition $Ba_{0.85}Ca_{0.15}Zr_{0.1}Ti_{0.9}O_3$ has the most uniform microstructure.

BCZT Piezoceramic Polarization Study

Heating ceramic samples to a glassy phase with subsequent cooling to room temperature leads to the fact that during cooling in the crystal cells a phase transition occurs, as a result of which the vectors of spontaneous piezoceramics polarization in the entire set of crystal lattices do not take the same direction, while regions (domains) appear.) having the same direction of spontaneous polarization [2]. In [3], $Ba_{0.85}Ca_{0.15}Zr_{0.1}Ti_{0.9}O_3$ (BCZT) ceramics were obtained using a conventional solid solution. Experimental studies of the dependences of the electrophysical parameters of the samples on temperature, electric field strength, and the surface morphology were studied using a scanning electron microscope. Based on the studies results [3], a graph of the dependence of the $Ba_{0.85}Ca_{0.15}Zr_{0.1}Ti_{0.9}O_3$ polarization on temperature and the strength of the external electric field was plotted (Fig. 1).

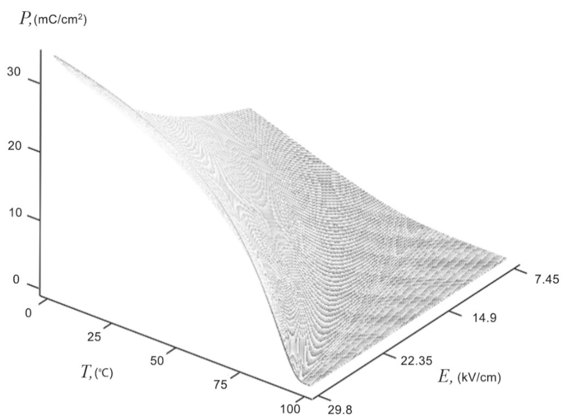

Fig. 1. Dependence of $Ba_{0.85}Ca_{0.15}Zr_{0.1}Ti_{0.9}O_3$ polarization P (mC/cm²) on temperature T (°C) and external electric field strength E (kV/cm)

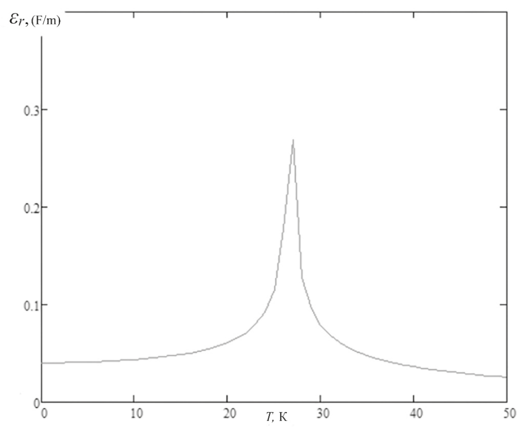

Fig. 2. Dependence of the $Ba_{0.85}Ca_{0.15}Zr_{0.1}Ti_{0.9}O_3$ complex permittivity on temperature

For a detailed study of the $Ba_{0.85}Ca_{0.15}Zr_{0.1}Ti_{0.9}O_3$ polarization, it is necessary to consider dielectric relaxation. In the case of dielectric relaxation, the polarization response to an external electric field is considered [4]. Dielectric relaxation is usually associated with the dynamics of electric dipoles of individual molecules or molecules groups, ions or electrons, passing between allowed energy levels. In the case of orientational polarization, the dipole moments of elementary dipoles remain constant in magnitude, but the directions change due to the thermal motion manifestation [5].

The activation energy of the dielectric relaxation was calculated using the Debye relation [6], the Debye relaxation can be expressed using the complex permittivity. The complex dielectric function is formed by summing the imaginary and real parts [7, 8], dependence of the imaginary part of the permittivity temperature [7]. The dependence of the $Ba_{0.85}Ca_{0.15}Zr_{0.1}Ti_{0.9}O_3$ complex permittivity on temperature is shown in Fig. 2.

To study the phenomenon of dielectric hysteresis of the polarization *P* dependence on the electric field strength *E*, the authors proposed an automated information-measuring system based on the modified Sawyer-Tower method [9, 10]. Fig. 3 shows the experimental dependence of the polarization of a ferroelectric ceramic sample based on barium titanate on the intensity electric field at a temperature of 80 °C.

The automated methods developed by the authors make it possible to estimate the coercive field (marked as “2” and “5” in Fig. 3) and the residual polarization (marked as “3” and “4” in Fig. 3, respectively) , as well as calculate the values of other electrophysical parameters, including the tangent of the dielectric loss angle, relative permittivity [11, 12].

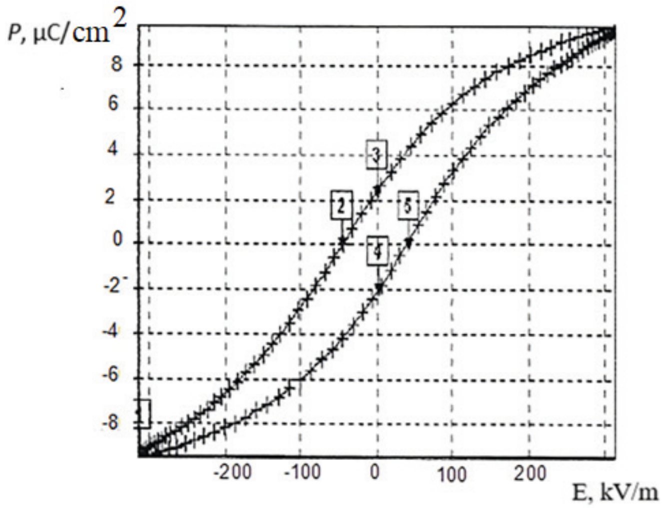

Fig. 3. Experimental dependence of the polarization of a ferroelectric ceramic sample based on barium titanate on the electric field strength at a temperature of 80 °C

Methods for measuring the tangent of the dielectric loss angle

To measure the dielectric loss tangent, the authors used two modifications of the Sawyer–Tower scheme (Fig. 4, *a*, *b*).

When using the first of the proposed methods, based on the scheme shown in Fig. 4,*a*, an indirect determination of the capacitance C_x is performed using the following formula:

$$c_x = \frac{U_y C_0}{U}, \tag{1}$$

where U_y is the voltage across a capacitor of known value , included in the lower arm of the capacitive divider.

Using scheme in Fig. 3,*b*, an indirect determination of r_x (the resistance) was made using the following formula:

$$r_x = \frac{\tau_1 r_1 - \tau_2 r_2}{\tau_2 - \tau_1}, \quad (2)$$

where r_1, r_2 are the known resistances of the resistors, which are connected in turn in series to the capacitor with the materials under study (Fig. 4,*b*); τ_1, τ_2 and r_1, r_2 are connected.

It should be noted that the relative measurement error is the sum of the relative measurement errors of frequency, capacitance and resistance:

$$\delta_1(tg\delta) = \delta f + \delta C_x + \delta r_x. \quad (3)$$

Next, we consider another method for indirectly measuring the dielectric loss tangent, which involves determining the area of the hysteresis loop of the dependence $U_y(U_{cx})$, where $U_{cx} = U - U_y = UC_0/(C_0 + Cx)$ is the voltage across the capacitor with the active dielectric under study.

The power W_a dissipated in the ferroelectric during the period of the sinusoidal signal T is directly proportional to the area of the hysteresis loop (see Fig. 3), $W_a = (1/T)\oint U_{cx}dq$, al under study $dq = C_0 dU_y$. Let us denote the integral in the numerator $J = \oint U_{cx} d\, U_y$, for its calculation it is advisable to use the methods of numerical integration. In this case, there is an error Δ_J, due, firstly, to the presence of a truncation error due to the replacement of curvilinear trapezoids by rectilinear ones and, secondly, to an error component caused by errors in measuring the values of the function $U_y(U_{cx})$.

To estimate the standard deviation of the instrumental error of indirect measurement under the normal distribution law, the expression is applicable:

$$\sigma_{tg\delta_2} = \pm\sqrt{\left(\frac{\partial tg\delta}{\partial J}\sigma_J\right)^2 + \left(\frac{\partial tg\delta}{\partial U_x}\sigma_{U_x}\right)^2 + \left(\frac{\partial tg\delta}{\partial U_y}\sigma_{U_y}\right)^2}, \quad (4)$$

where $J = \oint U_c\, dU_y$.

To calculate the ultimate absolute resulting error Δ_2, it is necessary to find the sum $\Delta_2 = \Delta_J + \Delta_{tg\delta 2}$, where $\Delta_{tg\delta 2} = 3.09\sigma_{tg\delta 2}$. In turn, in relative units, the measurement error of the dielectric loss tangent is determined by the expression:

$$\delta_2 = \frac{\Delta_2}{tg\delta} = \frac{\Delta_2 \pi (U_{C_x})_m (U_y)_m}{\oint U_{C_x} dU_y}. \quad (5)$$

The instrumental implementation of the Sawyer–Tower scheme modifications proposed by the authors makes it possible to measure the dielectric loss tangent with relative errors not exceeding, respectively, $\delta_1 \le 0.60\%$, $\delta_2 \le 0.65\%$.

Results and Discussion

As a result of studying the dependences of the electrophysical parameters of BCZT piezoceramics samples on temperature and electric field strength, graphs of the porosity dependence on influencing factors were obtained, which made it possible to study dielectric relaxation as a response of polarization to an external electric field. The relationship between the complex permittivity and temperature indicated a diffuse transformation confirming the relaxor-type behavior.

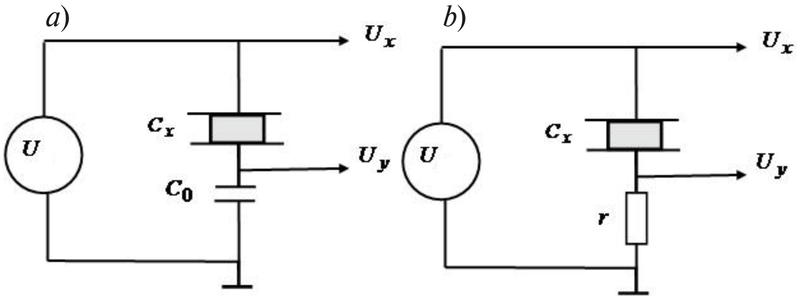

Fig. 4. Modified Sawyer-Tower circuits

Two modifications of the Sawyer–Tower scheme, the use of which is dictated by research goals: the hysteresis loops measurement (the scheme in Fig. 4,*a*) or the study of the repolarization of piezoceramic images, the measurement of the switching current versus time (the scheme in Fig. 4,*b*) are proposed. Both modifications make it possible to indirectly determine the parameters characterizing the dielectric hysteresis. Two methods for measuring the tangent of the dielectric loss angle based on metrological analysis are considered, formulas for estimating the limiting measurement errors, which in relative form do not exceed 0.60% and 0.65%, respectively, are obtained.

It is expedient to use the results presented in the study of ceramic piezos with ferroelectric properties, as well as in the design of functional electronics products based on active dielectrics.

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