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Reversible Nonlinearity, Opposite Piezoeffect, Electromechanical and Dielectric Hysteresis in "Ferroelectric-Soft" Multielement Compounds

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Abstract

The presented study focuses on the investigation of relationships between reverse piezomodule, half-cycles of electromechanical hysteresis loops, degree of polarization of specimens and amplitude of constant electric field strength, as well as polarization characteristics and concentration of components in solid solutions of the compound $(Pb_{1-\alpha_1-\alpha_2}Sr_{\alpha_1}Ba_{\alpha 2})\left[Ti_xZr_y\langle(Nb_{2/3}Zn_{1/3})(Nb_{2/3}Mg_{1/3})\rangle_{1-x-y}\right]O_3$. The results allowed to establish the correlation of polarization, deformation and structural properties of the studied solid solutions of the system. The study suggests the recommendations for practical application of the studied materials.

Keywords: Solid Solutions, Multicomponent Systems, Electromechanical Hysteresis, Deformation Characteristics.

Introduction

Nowadays, characteristics of piezoelectric materials must include relationships of their properties and external effects, in particular, strength of constant electric field (reversible nonlinearity, opposite piezoeffect, electromechanical and dielectric hysteresis) [1]. However, information on behavior of the opposite piezomodule d_{33}^{conv}, which acts as measure of deformation ξ_3 of the specimen in the direction of electric field directed its polar axis, in ferroelectric-soft (FS) materials with mobile domain structure is very scarce, which complicates evaluation of possibility of their application in positioning devices, where large values of shifts, which are induced by electric field, are required; that fact makes studies in that direction topical [2].

Objects, methods of production and study of specimens

The objects of the study were solid solutions (SS) of the compound $(Pb_{1-\alpha_1-\alpha_2}Sr_{\alpha_1}Ba_{\alpha 2})\left[Ti_xZr_y\langle(Nb_{2/3}Zn_{1/3})(Nb_{2/3}Mg_{1/3})\rangle_{1-x-y}\right]O_3$, where α_1 = 0. 02 − 0. 12, $\Delta\alpha_1$ = 0. 02, α_2 = 0. 0073 − 0. 045, x = 0. 395 − 0. 42, y = 0. 412 − 0. 437. The specimen were produced using regular ceramic technology (RCT), which includes two stages of synthesis ($T_{synt.\ 1}$ = 1140 K, τ = 5 h, $T_{synt.\ 2}$ = 1160 K, τ = 5 h; T_{sint} = 1570 K) with the following sintering without application of pressure [3]. All studies were carried out using non-polarized and polarized disc-shaped specimen of 10 mm diameter and 1 mm thickness.

X-Ray studies were carried out using powder X-Ray diffraction analysis method using DRON-3 diffractometer and powder dispersion analyzer (PDA) (FeK_α-radiation, Mn-filter; FeK_β-radiation; Bragg-Brentano focusing scheme).

The studies of relative reversible dielectric permeability $(\varepsilon_{33}^T/\varepsilon_0)_{rev}$ [5] of the objects were carried out using the laboratory bench, which was designed at the Physics Research Institute of Southern Federal University; the laboratory bench included a high-voltage rectifier (constant voltage source) for smooth supply of constant voltage to the studied specimen (from 0 to 4 kV), instruments measuring capacity and conductivity (AC bridge E-8-2, frequency 1 kHz), working chamber, where the studied specimen was placed, and the internal space of which was filled with polyethylene-siloxane liquid PES-5. In order to avoid interactions of the rectifier and the instrument measuring capacity, symmetrical circuit layout was used, which consists of two resistors, protecting the rectifier from short circuit in case of breakdown, and two capacitors, protecting the instrument measuring capacity from the influence of high voltage. Constant voltage was supplied via two resistors, which were connected in series with the studied specimen. The specimen was connected to the constant voltage source from one side; from another side it was serially connected through the capacitors to instrument measuring capacity. Capacity was measured after 5-10 min after each change of voltage at the specimen.

For measurements of longitudinal deformation ξ_3, which was induced by the electric field E applied to the piezoelement, we used the specially designed test bench. The test bench consisted of precision micrometer stand for mounting of piezoelement and instrument; stabilized source of voltage with smooth and discreet adjustment and digital indication of output voltage (voltage range 0-1550 V); galvanomagnetic dilatometer with digital indication of readings and capability to export them to PC or logger. In order to compensate temperature changes during measurements we used differential scheme of connection of induction sensors. The test bench allowed to measure relative deformation with accuracy of 10^{-5}. Calculation of reversible piezomodule was carried out using the following equation $d_{33}^{\text{conv.}} = \xi_3/E$ [6].

Loops of dielectric hysteresis were studied using oscilloscope method; the loops characterize relationship of polarization of dielectric and applied field; those relationships were used to identify full (P_f), induced (P_i), reorientation (P_r) and remanent (P_o) polarizations, as well as relationships of those values and coercive force and strength of electric field E.

Experimental Results and Discussion

Fig. 1 shows the relationship $(\varepsilon_{33}^{T}/\varepsilon_0)_{rev}$ of strength of constant displacing electric field E of SS of the discussed system. As it can be seen, all relationships are "butterfly"-shaped, which is specific for classical ferroelectrics [7]. Two characteristic areas of change can be marked out $(\varepsilon_{33}^{T}/\varepsilon_0)_{rev}$: first lies from α_1 = 0. 02 to α_1 = 0. 06 and has asymmetric loops, which strongly depend on α_1; second is located in the range 0. 06 ≤ α_1 ≤ 0. 12, and it has symmetric loops with large values $(\varepsilon_{33}^{T}/\varepsilon_0)_{rev_{\max}}$. Gradual increase of α_1 leads to narrowing of the loop, or, in other words, to gradual transition of ceramics into relaxor state, in which the relationship $(\varepsilon_{33}^{T}/\varepsilon_0)_{rev}$ has shape of dome [8], and increase of values of maximums of dielectric permeability with negative and positive values of strength of electric field.

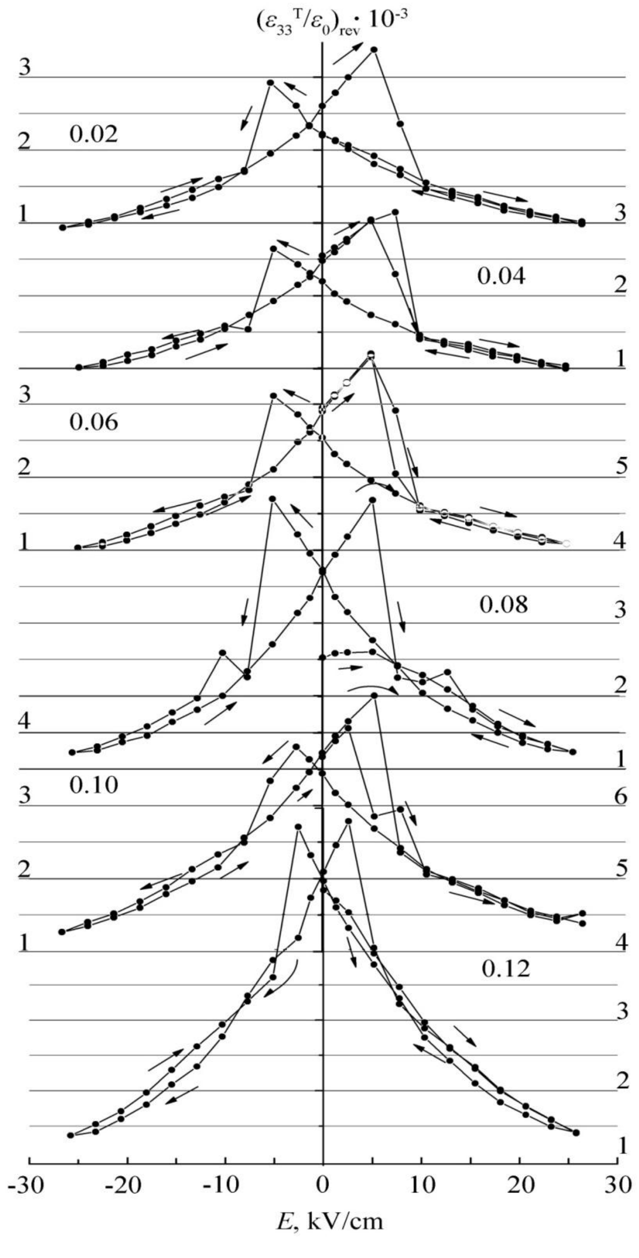

Figure 1: Relationship of reversible dielectric permeability of polarized specimens $\varepsilon_{33}{}^{T}/\varepsilon_0$ of the studied SS and strength of electric field (number near the curve represent concentration of Sr (α1) in arb. units).

High values of $(\varepsilon_{33}{}^{T}/\varepsilon_0)_{rev}$ and their strong dependence on value of E allow to attribute the studied ceramics to FS group [9]. That is supported also by concentration relationship of relative dielectric permeability. The latter is indicative of its increase in the studied ceramics with the increase of concentration of Sr (α_1) from values of $(\varepsilon_{33}^T/\varepsilon_0)_{rev_{max}}$, equal to 2500 for α_1 = 0. 02, to $(\varepsilon_{33}{}^T/\varepsilon_0)_{rev}$ = 6000 for α_1 = 0. 12. Values of $(\varepsilon_{33}^T/\varepsilon_0)_{rev_{max}}$ increase nonmonotonously with weak decrease with positive values of E and strong decrease for SS at α_1 = 0. 04 and α_1 = 0. 08 with negative zero values of E.

Fig. 2, *a* shows relationships of reversible piezomodule d_{33}^{conv} and half-cycles of loops of electromechanical hysteresis and amplitude of strength of constant electric field E of the studied SS.

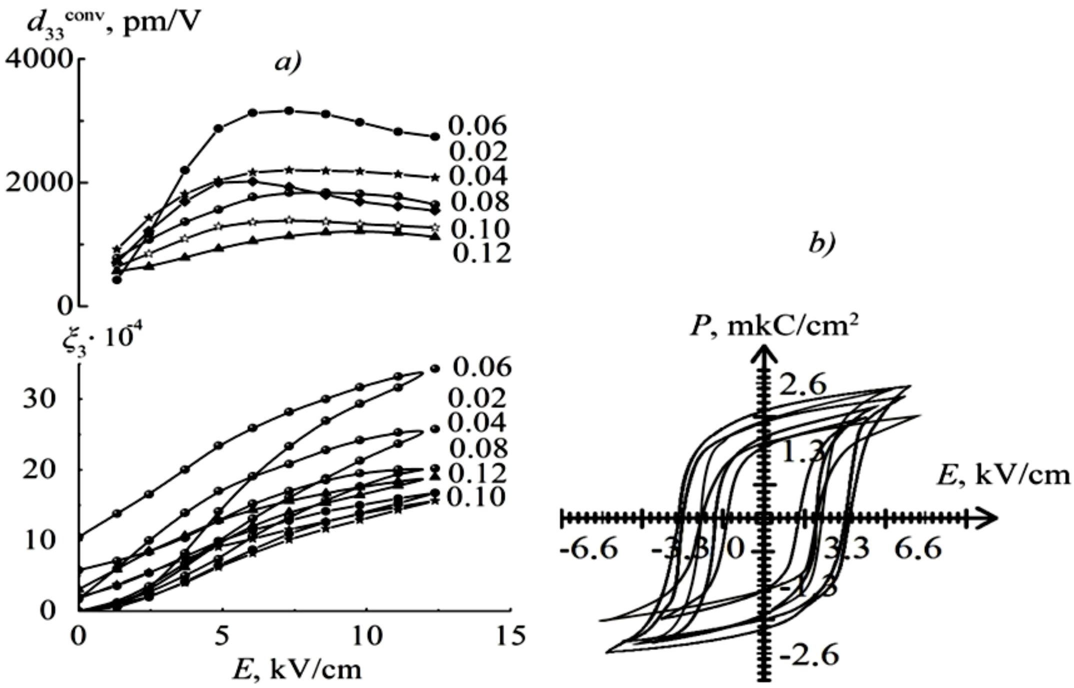

Figure 2: Relationship of reversible piezomodule d_{33}^{conv} and half-cycles of hysteresis (*a*), loops of dielectric hysteresis (*b*) and amplitude of strength of constant electric field E of the studied SS.

As it can be seen, in the studied fields' area all materials are characterized by monotonous relationships ξ_3 (E) both for ascending and descending half-cycles of loops of electromechanical hysteresis. Relationships of d_{33}^{conv} (E) are also of similar character: first, piezomodule increases with reaching of maximum at E = 6 kV/cm; them, a minor decrease occurs, which is specific for FS-ceramics. For ceramics at α_1 = 0. 06÷0. 12 there is more rapid increase of d_{33}^{conv}, which is, as it was mentioned in the study [10], specific for relaxor materials. Fig. 2, *b* demonstrates loops of dielectric hysteresis of the studied SS. The obtained ceramics has small values of polarization characteristics, which is indicative of the increase of ferroelectric-softness of the specimens.

Fig. 3 presents relationships d_{33max}, width of loop of electromechanical hysteresis $\Delta\xi_3$ at E = 5 kV/cm, permanent deformation ξ_3 and polarization characteristics and concentration of components in the studied system.

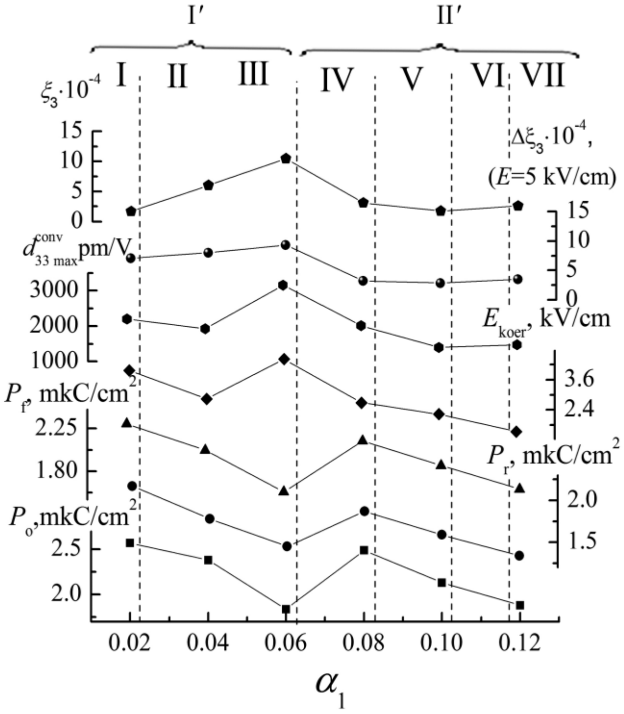

Figure 3: Relationships of d_{33max}^{conv}, width of loop of hysteresis $\Delta\xi_3$ at E = 5 kV/cm, permanent deformation ξ_3 and polarization characteristics of SS of the studied system and concentration of components. I' – areas, where tetragonal (T), rhombohedral (Rh) and pseudocubic (Psc) phases coexist, which are, depending on content of the mentioned phases, are specified by roman numerals I-III (I-70T+18Rh+12Psc; II-75T+15Rh+10Psc; III-75T+15Rh+10Psc). Arabic numerals before names of phases represent their composition, in %. Same for the area II' with coexisting T and Rh phases (IV-80Ti+20Rh; V-80T+20Rh; VI-75T+25Rh; VII-80T+20Rh). Ti (I = 1, 2) – phase conditions inside T field with single symmetry, which differ in cell parameters [11].

Analysis of Fig. 3 shows good correlation of polarization, deformation and structural properties of the studied SS system: maximum values of d_{33}^{conv} and ξ_3, as well as decreased polarization characteristics correspond to the zone of transition form three-phase to two-phase condition.

On the basis of the obtained data the materials with d_{33}^{conv} = 3000 pm/V, 0. 65 $\leq K_p \leq$ 0. 70, $T_C \approx (420 - 570)$ K (see Table) can be marked out, which are prospective from the point of view of development of high-voltage actuators, laser adaptive systems, vibration compensators and precise positioning systems.

Table 1: Electrophysical parameters of piezoceramic materials of the studied SS.

No.	$\varepsilon^T_{33}/\varepsilon_0$	K_p	$\lvert d_{31}\rvert$, pC/N	d_{33}, pC/N	d_{33}^{conv}, pm/V ($E = 1$ (kV/cm))	$Y^E_{11}\cdot 10^{-11}$, N/m^2	$V^E_1 \cdot 10^3$, m/s	$tg\delta$, % (E = 50 V/cm)	$\lvert g_{31}\rvert$, mV·m/N	Q_M
1	2700	0.70	235	540	900	0.551	2.681	1.7	9.9	75
2	3020	0.65	238	547	670	0.587	2.792	1.26	8.8	69

Conclusion

In the presented study polarization, deformation and structural characteristics of solid solution of the system $(Pb_{1-\alpha_1-\alpha_2}Sr_{\alpha_1}Ba_{\alpha 2})\left[Ti_xZr_y\langle(Nb_{2/3}Zn_{1/3})(Nb_{2/3}Mg_{1/3})\rangle_{1-x-y}\right]O_3$ were studied. Also, possibility of practical application of the studied materials in piezo-devices was discussed.

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