Features of Dielectric and Electromechanical Response of PMN-PT Based Materials in Electric Fields

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Abstract: In this paper, the characteristics of the dielectric and electromechanical response of ceramics mPbMg\(_{1/3}\)Nb\(_{2/3}\)O\(_3\)-nPbNi\(_{1/3}\)Nb\(_{2/3}\)O\(_3\)-yPbZn\(_{1/3}\)Nb\(_{2/3}\)O\(_3\)-xPbTiO\(_3\) (m = 0.1298-0.4844, n = 0.1266-0.4326, y = 0.0842-0.130, x = 0.25-0.40)-analogue of a class of materials based on relaxor ferroelectrics and PbTiO\(_3\), characterized by the presence of morphotropic phase boundary have been studied. It is shown the evolution of \(\varepsilon/\varepsilon_0(E)\) and \(\zeta_3(E)\)-dependence of the studied ceramics with increasing content of x from typical relaxor ferroelectrics up to the classic ferroelectrics. The existence of a plateau-like anomalies on \(\varepsilon/\varepsilon_0(E)\) and \(\zeta_3(E)\)-dependency of heterophase ceramics (x \(\approx\) 0.30-0.35) has been established. It is shown that the cause of these anomalies is termination of non-180°- domain switching due to high mechanical stresses which arise because of E-field induced phase transition from heterophase (Psc + T) to single-phase (T) state. It is found that ceramics with x = 0.275 have the best parameters for the application in tunable devices and actuators. The existence of a plateau-like anomaly on the \(\varepsilon/\varepsilon_0, K\) and \(\zeta_3\) dependeces of E of ceramics near the morphotropic phase boundary is an extremely important feature for practical applications, which should be taken into account when creating a high-performance actuators and tunable devices.

Key words: Relaxor ferroelectrics, morphotropic phase boundary, dielectric permittivity, longitudinal strain, tunability, electric field induced phase transition.

Nomenclature

| RF: Relaxor ferroelectric | \(\zeta_3\): Longitudinal strain |
| E: Electric field | \(\varepsilon/\varepsilon_0\): Relative dielectric permittivity |
| K: Tunability | \(\zeta_{3,max}\): Maximal longitudinal strain |
| MPB: Morphotropic phase boundary | \(\zeta_{3, rem}\): Remanent longitudinal strain |
| SS: Solid solution | \(a_T\): Tetragonal unit cell parameter |
| T: Tetragonal phase | \(c_T\): Tetragonal unit cell parameter |
| Rh: Rhombohedral phase | \(a_{Psc}\): Pseudocubic unit cell parameter |
| M: Monoclinic phase | \(V_T\): The volume of the tetragonal unit cell |
| O: Orthorhombic phase | \(E_c\): Coercive field |

Greek Letters

1. Introduction

Dependences of the longitudinal strain \(\zeta_3\) and relative permittivity \(\varepsilon/\varepsilon_0\) on the electric field (E) magnitude are the most important characteristics of materials for applications in electro-mechanical actuators, and tunable devices. On the basis of the latter it is possible to determine the tunability coefficient: \(K = \left[\left(\varepsilon/\varepsilon_0 (E = 0) - \varepsilon/\varepsilon_0 (E \neq 0)\right)/\varepsilon/\varepsilon_0 (E = 0)\right]\times100\%\), one of the main functional parameters of the materials for use in the microwave range devices, such as radar and communication systems [1, 2].

For these practical applications materials are promising based on solid solutions (SS) of relaxor
ferroelectrics (RF) PbMg\(_{1/3}\)Nb\(_{2/3}\)O\(_3\) (PMN), PbZn\(_{1/3}\)Nb\(_{2/3}\)O\(_3\) (PZN), PbNi\(_{1/3}\)Nb\(_{2/3}\)O\(_3\) (PNN) and classical ferroelectric PbTiO\(_3\) (PT). Single crystals of these compositions near the morphotropic phase boundary (MPB), separating the tetragonal (T) and rhombohedral (Rh) areas of the phase diagram, show the record values of the piezoelectric response (\(d_{33} > 2,500\) pC/N), the electromechanical coupling coefficient (\(k_{33} > 0.90\)) and high \(\varepsilon/\varepsilon_0\) [3].

One feature of \(\varepsilon/\varepsilon_0\) (E)-dependence of ferroelectric materials is the presence of minima which appear with increasing E (after changing the E direction). For the first time these anomalies were found by Drougard and Young in 1954 in the study of T-BaTiO\(_3\) crystal [4]. Their formation was attributed to mechanical clamping of the antiparallel domains, the number of which is maximum near the coercive fields \(E_c\). Similar anomalies were observed in the study of \(\varepsilon/\varepsilon_0\) (E), dependences of the orthorhombic (O) BaTiO\(_3\) crystal [5], ceramics based on the system (1-x)PbZrO\(_3\)-xPbTiO\(_3\) (PZT) [6-10], (011)-oriented crystal (1-x)PbMg\(_{1/3}\)Nb\(_{2/3}\)O\(_3\) (PMN)-xPbTiO\(_3\) (PT) (x = 0.32) [11], and lead-free ceramics based on the system (1-x)(Na, K, Li, Ba)(Nb\(_{0.9}\)Ta\(_{0.1}\))O\(_3\)-xSrZrO\(_3\) [12].

In Ref. [13], we showed the evolution of \(\varepsilon/\varepsilon_0\) (E)-dependency of multicomponent system PMN-PNN-PZN-PT ceramics at a concentration transition from classical ferroelectric to RF. With a decrease of PT transformation, typical for classical ferroelectrics, minima of \(\varepsilon/\varepsilon_0\) in plateau-like anomalies are detected. This feature was associated with the termination of the non-180\(^\circ\)-domain switching due to high mechanical stresses caused by E induced phase transition from the heterophase (pseudocubic (Psc) phase (symmetry which, because of the weakness and blur X-ray peaks was not possible to identify) + T-phase) in single-phase (T-phase) state [14].

There are several models to explain the appearance of minima on the \(\varepsilon/\varepsilon_0\) (E) Rh, T and heterophase ceramics based on contributions to \(\varepsilon/\varepsilon_0\) switching of 180\(^\circ\) and 90\(^\circ\) (in the case of T samples) and 71\(^\circ\) and 109\(^\circ\) (in the case of Rh samples) domains [6, 9]. However, these models cannot explain the formation of plateau-like anomalies on \(\varepsilon/\varepsilon_0\) (E)-dependencies of heterophase ceramics [13, 14].

Besides the switching processes under the influence of E, induced phase transitions, leading to significant changes in the behavior of macroscopic properties, can occur. Thus, the appearance of a plateau on \(\varepsilon/\varepsilon_0\) (E)-dependencies of (001)-oriented crystal (1-x) PZN - x PT (x = 0.08) [15] was associated with an electric field induced phase transition in the T-phase, the presence of which was confirmed later in Ref. [16]. This increase in E in the case of compositions near MPB (1-x)PMN-xPT (x ≈ 0.27-0.32) and (1-x)PZN-xPT (x ≈ 0.07-0.09), results in a complicated sequence of phase transitions [17, 18]. On the basis of ab initio calculations Fu and Cohen showed that the source of the giant piezoelectric effect in these materials may be polarization-rotating mechanism associated with the rotation of the polarization vector via phase transitions from Rh to the T-phase through an intermediate phase(s) [19]. Vanderbilt and Cohen, using the Landau-Ginzburg-Devonshire potential of the 8th order, predicted the existence of two different monoclinic (M) phases with space groups Cm (M\(_A\), M\(_B\)) and Pm (M\(_C\)) [20], which together with the O-phase may act as a structural bridge at E induced phase transitions [21]. These phases were observed in the study of PMN-PT [22, 23], PZN-PT [24, 25] and PZT [26, 27] near MPB. In addition, some studies have shown that near MPB several phases the mentioned above can coexist [28, 29]. They lead to the complexity of the domain structure [30, 31], and as a result, affect the manifestation of the macroscopic properties.

However, the binary system (for example based on RF and PT) is characterized by one-dimensional MPB. This reduces the variability of the functional SS, which limits the choice of materials based on these systems with the required combinations of parameters. In addition, MPB may represent a very narrow range of concentrations of the components, which complicates
the study of SS near it. One way to solve these problems is to construct multi-component compositions [32, 33]. Increasing the number of components allows us to design systems with multi-dimensional MPB. The latter enables to increase significantly the range of concentrations of the existence of MPB. In the same way using of this approach significantly expands the choice of the SS with specified combinations of parameters.

In this paper, we consider the features of $\varepsilon/\varepsilon_0$ (E) and $\zeta_3$ (E)-dependences of multi-component ceramics based on RF and PT. The combined examination of these characteristics allows us to make conclusions about the possible causes of the observed anomalies. Special attention is paid to the aspects of practical application of materials based on the RF and PT near MPB.

2. Experiments

The ceramics of system $m$PMN-$n$PNN-$y$PZN-$x$PT ($m = 0.1298-0.4844$, $n = 0.1266-0.4326$, $y = 0.0842-0.130$, $x = 0.25-0.40$), modified by 5 mol. % of barium were prepared by conventional solid state synthesis with the help of columbite precursor route [34]. Synthesis of columbite-like compositions MgNb$_2$O$_6$, NiNb$_2$O$_6$, ZnNb$_2$O$_6$ from oxides MgO, NiO, ZnO and Nb$_2$O$_5$ included two stages: for ZnNb$_2$O$_6$ and MgNb$_2$O$_6$ they were synthesized at temperatures $T_1 = 1,000 \, ^\circ\text{C}$, $T_2 = 1,100 \, ^\circ\text{C}$ during 6 and 4 h, respectively; for NiNb$_2$O$_6$ temperatures of synthesis were $T_1 = 1,000 \, ^\circ\text{C}$, $T_2 = 1,240 \, ^\circ\text{C}$ during 6 and 2 h, respectively. SS of a final composition were produced by a single-phase synthesis of columbite composition that were obtained earlier and also by the synthesis of PbO, TiO$_2$ and BaCO$_3$ (in case of modified samples) at $T = 950 \, ^\circ\text{C}$ during 4 h. The choice of the studied compounds and MPB prediction were described in detail in Ref. [35]. In addition, the optimal conditions (synthesis and sintering regimes) for the preparation of studied SS were determined. Sintering was carried out at $T_{\text{sint.}} = 1,180 \, ^\circ\text{C}$. The samples were characterized by a high experimental density (95% of theoretical) and there were no traces of pyrochlore phase on X-ray diffraction patterns.

Samples were represented by discs with the 10 mm diameter and 1 mm thickness with silver-bearing electrodes applied (by double firing) to the flat butt end surfaces.

Measurement of $\varepsilon/\varepsilon_0$ (E) dependences was made at the magnitude and frequency of the measuring electric field 1 V and 1 kHz, respectively, on the testing bench which includes LCR-meter Agilent 4263B. DC voltage of shifting electric field was changing reversely in the range -15 – (+15) kV/cm. A specially designed stand was used to measure the longitudinal strain $\xi_3$ induced by discretely increasing or discretely decreasing the electric field applied to samples. The main stand element is a galvanomagnetic dilatometer with a digital indication of readings and the possibility of displaying them on a recorder and a computer. [36]. Measurements were made in quasi-static mode range of -15 – (+15) kV/cm.

3. Results and Discussion

Fig. 1 shows $\varepsilon/\varepsilon_0$ (E)-dependences of studied ceramics with various concentrations of PT ($x$). It can be seen that the samples with a high concentration of PT ($x = 0.40$) is characterized by $\varepsilon/\varepsilon_0$ (E)-dependences typical for classical ferroelectrics. The recession of $\varepsilon/\varepsilon_0$ curve is associated with switching of non-180° domains, enlargement of the domain structure, and, as a consequence, the decrease of the contribution the domain walls motion. Minimum of $\varepsilon/\varepsilon_0$ which is observed at $E \approx 7.0 \, \text{kV/cm}$, is due to antiparallel domain clamping effect [4]. The decrease of $E$ leads to a monotonic increase of $\varepsilon/\varepsilon_0$, with hysteresis between forward and reverse moves of $\varepsilon/\varepsilon_0$ (E). The decrease in concentration of PT ($x < 0.40$) leads to the transformation of $\varepsilon/\varepsilon_0$ minimum in plateau-like sections ($x = 0.30-0.325$) and their disappearance when $x < 0.30$. Moreover, with decreasing of PT concentration morphotropic phase transition from T to
cubic (C) phase occurs [35, 37]. There is a region of coexistence of T and Psc phases at $x \approx 0.30$-0.375. Samples with $x \approx 0.275$ are characterized by Psc state.

Fig. 2 shows $\zeta_3(E)$ dependences of studied ceramics with various PT($x$) concentrations. With a high content of PT ($x \geq 0.35$), typical for classical ferroelectrics “butterfly”-like curves are observed. The decrease in concentration of PT ($x < 0.325$) leads to the evolution of $\zeta_3(E)$-form to the typical for RF (“sprout” curves [38]). They are characterized by absence of hysteresis between forward and reverse motion of $\zeta_3(E)$-curve. It is an important advantage in using such materials in actuators.

On $\zeta_3(E)$-dependences of heterophase samples with $x = 0.30$ and 0.325 in the direct course of E at ($E \approx 4$- 6 kV/cm) plateau-like anomaly is observed. The manifestation of such anomaly on $\varepsilon/\varepsilon_0$ (E)-dependence was associated in [14] with the termination of the non-180°-domain switching due to high mechanical stresses caused by E induced phase transition from heterophase (Psc + T) to single-phase (T) state. Since the contribution to $\zeta_3$ is made by only non-180° domain switching, the appearance of a plateau on $\zeta_3(E)$-dependence is quite logical. Thus, in the range of $E = 3.5$-7.5 kV/cm there are areas in which $\varepsilon/\varepsilon_0$ and $\zeta_3$ of studied heterophase ceramics near the -MPB ($x \approx 0.30-0.35$) are practically independent on E.

Fig. 3 shows a comparison of $\varepsilon/\varepsilon_0$, $\zeta_3$, K and the unit cell parameters, obtained by X-ray diffraction measurement in [14], of heterophase sample with $x = 0.30$ from value E. There is a sharp increase of T cell parameter $c_T$ (deformation along the direction of E) and the volume of the T cell ($V_T$) in the field of E, corresponding to the plateau on the curves of $\zeta_3(E)$, $\varepsilon/\varepsilon_0$ (E) and K (E). In addition, at $E \approx 5$ kV / cm induced phase transition from heterophase (T + Psc) to single-phase state (T) occurs, accompanied by a termination of non 180° domain switching in the range $E = 4.5$-6.3 kV/cm [14]. We assume that the termination of domain switching can be caused by
Fig. 3 Depending on the parameters ($a_{pm}$, $a_{c}$, and $c_{1}$), volume ($V_{f}$) unit cell, $\varepsilon/\varepsilon_{0}$, K and $\zeta_{3}$ of heterophase sample with $x = 0.30$ on E [14].

mechanical stresses during induced phase transition. The existence of a plateau on the $\varepsilon/\varepsilon_{0}(E)$, K(E) and $\zeta_{3}$ (E) of ceramics near the MPB is an extremely important feature for practical applications, since these materials are characterized by the highest piezoelectric responses [3]. The presence of this anomaly should be considered when creating high-performance actuators and tunable devices based on SS of RF and PT.

Fig. 4 shows a diagram that displays the dependence of K (at E = 20 kV/cm) and $\varepsilon/\varepsilon_{0}$ (at E = 0) on the concentration of PT (x). Highlighted areas of PT-concentration, which correspond to the most common $\varepsilon/\varepsilon_{0}$ (E)-dependence, are characterized by:

1) High values of K (~80%) (they almost independent of x), and $\varepsilon/\varepsilon_{0}$ (~10.000), hysteresis-free $\varepsilon/\varepsilon_{0}$ (E) dependence, absence of pronounced anomalies of $\varepsilon/\varepsilon_{0}$ (region I, x < 0.30);

2) Mean values of K (35%-80%) and $\varepsilon/\varepsilon_{0}$ (3,000-8,000), hysteresis of $\varepsilon/\varepsilon_{0}$ (E)-dependence, appearance of plateau-like anomalies and implicit minima of $\varepsilon/\varepsilon_{0}$ with increasing E (region II, 0.30 ≤ x < 0.375);

3) Low values of K (~30%) and $\varepsilon/\varepsilon_{0}$ (2,500-3,000), a pronounced hysteresis of $\varepsilon/\varepsilon_{0}$ (E)-dependencies, the formation of explicit minima of $\varepsilon/\varepsilon_{0}$ with increasing E (region III, x ≥ 0.375).

Chart analysis showed that ceramics of group I have the best parameters for application in tunable devices. The ceramics application of groups II and III in such devices can be complicated by the low values of K, $\varepsilon/\varepsilon_{0}$ and the presence of anomalies in the $\varepsilon/\varepsilon_{0}$ (E)-dependencies. At the same time SS of group III can be used in devices that are resistant to external influences (E) due to their low K.

Fig. 5 shows a diagram that displays the dependence of the maximum ($\zeta_{3,\text{max}}$) and remanent ($\zeta_{3,\text{rem}}$) deformation on the concentration of PT (x). Curve $\zeta_{3,\text{max}}(x)$ is characterized by two maxima at x = 0.275 and $x = 0.35-0.325$ The first maximum, probably is due to the fact that the Psc phase may be M symmetry. It is a well-known fact that M-SS based on the RF and PT have high values of E induced strains. This can be explained by a contribution of the polarization rotation mechanism during successive induced phase transitions [21], and a large number (24) of possible domain configurations. The appearance of the second maximum turns out to be associated with the transition to heterophase (T + Psc) state with increasing x. As a result, the number of possible
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Fig. 5  Diagram showing the dependence of the maximum ($\zeta_3, \text{max}$) (full symbols) and remanent ($\zeta_3, \text{rem}$) (open symbols) strain on the concentration of PT ($x$).

domain configurations is 30 (24–M + 6–T domains). Curve $\zeta_3, \text{rem} (x)$ is characterized by a maximum at $x = 0.35$. Perhaps its appearance on $\zeta_3, \text{rem} (x)$-dependence is caused by the mutual action of the two processes. As is well-known, an increase in the content of lead titanate in materials based on RF and PT leads to the stabilization of the ferroelectric state and the formation of the normal domain structure. As a result, the values of $\zeta_3, \text{rem}$ rise. However, an increase in $x$ also increases the coercive field, which affects the amount of switched domains and reduces $\zeta_3, \text{rem}$. The superposition of these two processes may give rise to a maximum on $\zeta_3, \text{rem} (x)$-dependence.

For applications in actuators, as well as in the tunable devices, the best parameters are established in ceramics of group I (in particular, $x = 0.275$). These SS are characterized by high values of $\zeta_3, \text{max}$, weak hysteresis of $\zeta_3(E)$-dependence and absence of pronounced $\zeta_3$ anomalies. Ceramics of group II may be used in sensors of pressure, deformation, etc. due to their high piezoelectric coefficient [35, 37]. Materials of group III can be used in force piezoelectric devices, such as piezomotors.

4. Conclusions

In this paper, the features of the dielectric and electromechanical response of ceramics $\text{mPMN} - \text{nPNN-yPZN-xPT}$ ($m = 0.1298-0.4844$, $n = 0.1266-0.4326$, $y = 0.0842-0.130$, $x = 0.25-0.40$), which is analogue of a class of RF and PT based materials, characterized by the presence of MPB, have been studied. It is shown the evolution of $\varepsilon/\varepsilon_0 (E)$ and $\zeta_3 (E)$-dependences of the studied ceramics with increasing of PT content from typical for the RF to normal ferroelectrics. The existence of plateau on $\varepsilon/\varepsilon_0 (E)$ and $\zeta_3 (E)$-dependences of heterophase ceramics ($x \approx 0.30-0.35$) has been established. It is shown that the cause of these anomalies is termination of not 180°-domain switching due to high mechanical stresses caused by $E$ induced phase transition from the heterophase (Psc + T) to single-phase (T) state. Diagrams showing the value of the function parameters $K$, $\varepsilon/\varepsilon_0$, $\zeta_3, \text{max}$ and $\zeta_3, \text{rem}$ of studied ceramics on the concentration PT are constructed. Areas of PT concentration are highlighted, which correspond to the most common $\varepsilon/\varepsilon_0 (E)$ and $\zeta_3 (E)$-dependence. It is established that ceramics with $x = 0.275$ have the best parameters for the application in tunable devices and actuators. The existence of a plateau on the $\varepsilon/\varepsilon_0 (E)$, $K(E)$ and $\zeta_3(E)$ dependencies of SS near MPB is an extremely important feature for practical applications.

The presence of this anomaly should be taken into account when creating a high-performance actuators and tunable devices based on SS of RF and PT.

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