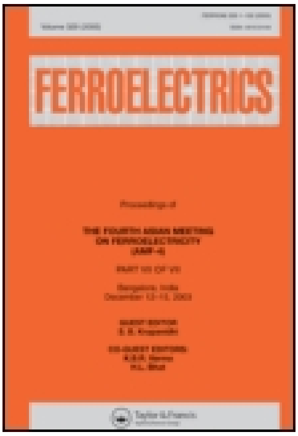

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Electromechanical Coupling Factors of Novel 0–3–0 Composites Based on PMN–*x*PT Single Crystals

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This paper is devoted to the problem of the electromechanical coupling of advanced composites based on relaxor-ferroelectric single crystals with large piezoelectric activity. A novel 0–3–0 PMN–0.33PT single crystal/porous araldite composite is considered as a material with large values of electromechanical coupling factors with a variable anisotropy. The role of physical and microstructural factors in achieving the considerable electromechanical coupling is discussed for 0–3–0 connectivity.

Keywords Composite; Relaxor-ferroelectric single crystal; Porous polymer; Electromechanical coupling factor

Single crystals (SCs) of relaxor-ferroelectric solid solutions of

$$(1-x)\mathrm{Pb}(\mathrm{Mg}_{1/3}\mathrm{Nb}_{2/3})\mathrm{O}_3 - x\mathrm{PbTiO}_3(\mathrm{PMN}-x\mathrm{PT})$$

with perovskite-type structure are of interest as highly effective components of advanced piezo-active composites. Electromechanical coupling is one of the main characteristics of any piezoelectric media, including composites with the piezo-active components. Recent results on the laminar [1, 2], matrix cellular [3] and fibrous [4] composites based on PMN–0.33PT SCs demonstrate the key role of the piezoelectric coefficients d_{ij} of SC in achieving considerable electromechanical coupling in these composites. Particular features of the behaviour of the electromechanical coupling factors (ECFs) in the 0–3 PMN–*x*PT SC/polymer composites were discussed in work [3], although the influence of the physical properties of the polymer matrix on different ECFs has not been considered in detail. The aim of the present study is to analyse ECFs in PMN–*x*PT/porous polymer composites with 0–3–0 connectivity.

The determination of the effective electromechanical properties of the 0–3–0 composite (Fig. 1) is carried out within the framework of the model of the piezo-active spheroidal inclusion in the matrix [3, 4]. It is assumed that the SC inclusions are characterised by equal sizes and a regular distribution in the matrix. The shape of each SC inclusion is described

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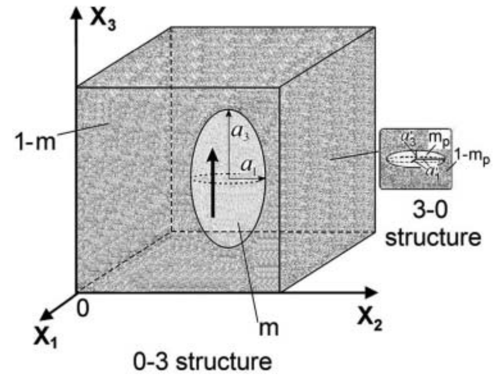

Figure 1. Schematic of the 0–3–0 SC/porous polymer composite. $(X_1X_2X_3)$ is the rectangular co-ordinate system. m and $1 - m$ are volume fractions of SC and porous polymer, respectively, and the spontaneous polarisation vector of SC is shown by arrow. In the porous polymer matrix, m_p and $1 - m_p$ are volume fractions of air pores and surrounding polymer, respectively. a_i and a_i' are semi-axes ($i = 1$ and 3) of the SC inclusion and pore, respectively.

by the equation $(x_1/a_1)^2 + (x_2/a_1)^2 + (x_3/a_3)^2 = 1$ relative to the axes of the rectangular co-ordinate system $(X_1X_2X_3)$, where the semi-axes of the spheroid are $a_1 = a_2$ and a_3, and $\rho = a_1/a_3$ is the aspect ratio. The main crystallographic axes (x, y, z) in each SC inclusion are oriented as follows: x ∥ OX_1, y ∥ OX_2 and z ∥ OX_3, and the spontaneous polarisation vector is oriented along the OX_3 axis (Fig. 1). It is assumed that the centres of symmetry of the inclusions occupy sites of a simple lattice with unit-cell vectors parallel to the OX_j axes. The matrix surrounding the inclusions contains spheroidal air pores (3–0 connectivity, Fig. 1) with an aspect ratio $\rho_p = a_1'/a_3'$ and semi-axes a_i'. The pores are considered to be regularly distributed in the polymer medium and the linear dimensions of each pore are much less than those of the SC inclusion. The effective electromechanical properties of the 0–3–0 composite are determined by means of the finite element method (FEM) [5] and effective medium method (EMM) [4]. The FEM and EMM are often used to predict the effective electromechanical properties and related parameters of piezo-active composites with appointed microgeometry.

A 9 × 9 matrix characterising the effective properties of the 0–3–0 composite studied is determined within the framework of the long-wave approximation [4] and given by

$$\|C^*\| = \|C^*(m,\ m_p,\ \rho,\ \rho_p)\| = \begin{pmatrix} ||c^{*E}|| & ||e^*||^t \\ ||e^*|| & -||\varepsilon^{*\xi}|| \end{pmatrix}, \tag{1}$$

where the superscript "t" denotes the transposition, $\|c^{*E}\|$, $\|e^*\|$ and $\|\varepsilon^{*\xi}\|$ are matrices of elastic moduli (at electric field E = const), piezoelectric coefficients and dielectric permittivities (at mechanical strain ξ = const), respectively. Based on the full set of electromechanical constants from Eq. (1) and taking into account conventional formulae for the piezoelectric medium [4, 6], we calculate the following ECFs of the composite: $k_{33}^* = d_{33}^*/(\varepsilon_{33}^{*\sigma} s_{33}^{*E})^{-1/2}$ (longitudinal ECF), $k_{31}^* = d_{31}^*/(\varepsilon_{33}^{*\sigma} s_{11}^{*E})^{-1/2}$ (lateral ECF), $k_t^* = e_{33}^*/(\varepsilon_{33}^{*\xi} c_{33}^{*D})^{-1/2}$ (thickness ECF), $k_p^* = k_{31}^*[2s_{11}^{*E}/(s_{11}^{*E} + s_{12}^{*E})]^{-1/2}$ (planar ECF), and $k_h^* = d_h^*/(\varepsilon_{33}^{*\sigma} s_h^{*E})^{-1/2}$ (hydrostatic ECF), where s_{ab}^{*E} is elastic compliance at E = const and $s_h^{*E} = 2(s_{11}^{*E} + s_{12}^{*E} + 2s_{13}^{*E}) + s_{33}^{*E}$ is hydrostatic elastic compliance at E = const. The ECFs characterise an effectiveness of the conversion of mechanical energy into electric one and vice versa at different oscillation modes of a piezoelectric element [6].

To calculate || C^* || from Eq. (1) and subsequently the ECFs of the composite, we use experimental room-temperature electromechanical constants of [001]-poled domain-engineered PMN–0.33PT SC (macroscopic 4*mm* symmetry) [7] and araldite [8]. In an attempt to attain large absolute values of the piezoelectric coefficients of the composite and to weaken the depolarising electric field in the presence of SC inclusions, we consider composite structures with the prolate SC inclusions ($0 < \rho < 1$, see Fig. 1). In addition, the oblate shape of the polymer pores ($\rho_p > 1$) leads to considerable elastic anisotropy of the polymer matrix and a favourable re-distribution of internal electric and mechanical fields in the composite structure. Results on ECFs predicted at $\rho = 0.1$ and $\rho_p = 100$ are shown in Fig. 2. Graphs in Fig. 2 demonstrate the potential advantages concerned with large values of ECFs and their anisotropy in the wide volume-fraction (m) range. The increase in

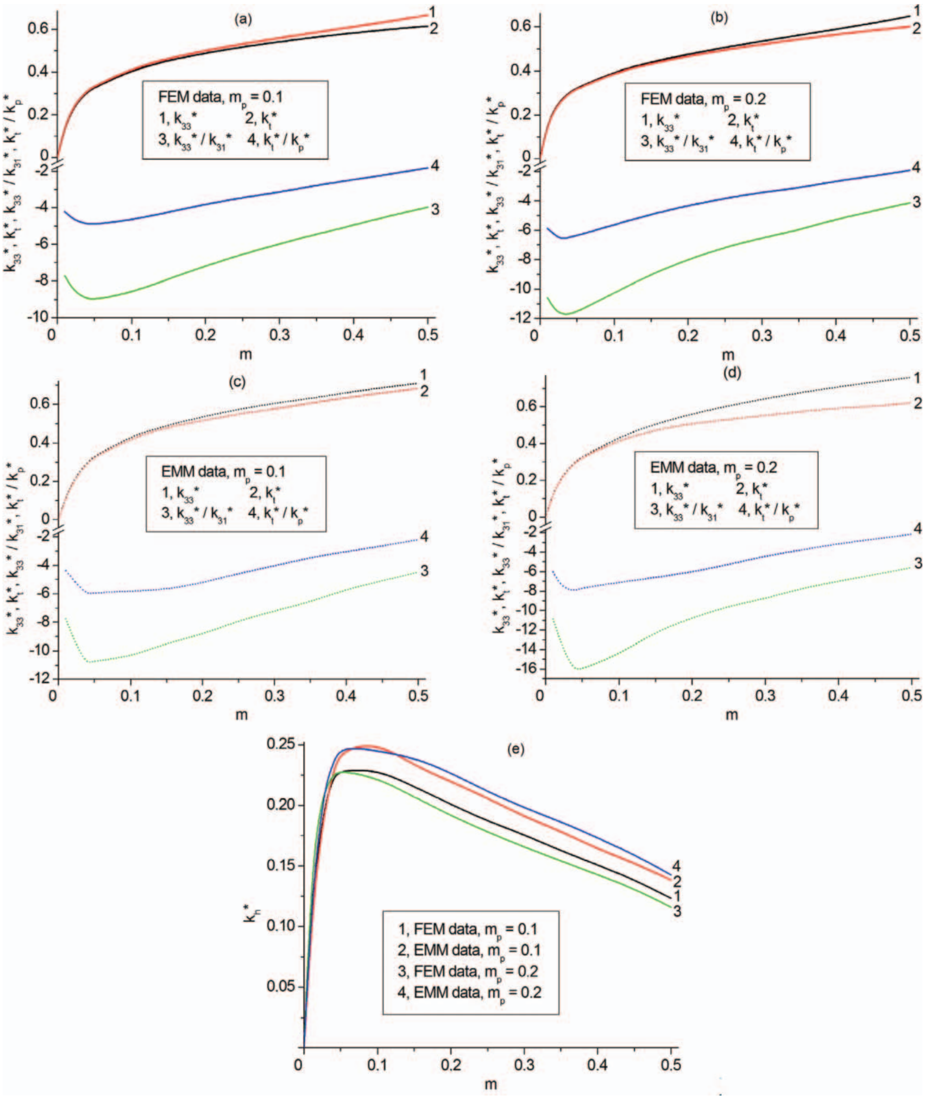

Figure 2. Volume-fraction (m) dependence of ECFs and their anisotropy in the 0–3–0 PMN–0.33PT SC/porous araldite composite at $\rho = 0.1$, $\rho_p = 100$ and $m_p = 0.1$–0.2. Calculations were made by means of the FEM (graphs a, b and e) and EFM (graphs c, d and e).

the porosity m_p of the matrix surrounding the piezo-active inclusions leads to an increase in the anisotropy of ECFs k_{33}^*/k_{31}^* and k_t^*/k_p^* (Fig. 2a–d) due to an increase in the elastic anisotropy of the porous matrix. The non-monotonic volume-fraction dependence of k_h^* (Fig. 2e) is accounted for by max d_h^* (due to the presence of the prolate SC inclusions and sgn $d_{33}^* = -$sgn $d_{31}^* > 0$ in the composite), relatively slow increasing $\varepsilon_{33}^{*\sigma}$ and decreasing s_h^{*E} with an increase of the volume fraction m. The ratios of k_{33}^*/k_{31}^* and k_t^*/k_p^* near the minimum points (Fig. 2a–d) are comparable to those in anisotropic ferroelectric ceramics of modified $PbTiO_3$ (see, e.g., data in Ref.9), however, in the wide m range, the values of ECFs k_{33}^* and k_t^* remain considerably larger than those of the aforementioned ceramics. As for the PMN–0.33PT SC, its ECFs k_{33} and k_t equal 0.94 and 0.64, respectively, and ratios of $k_{33}/k_{31} = -1.59$ and $k_t/k_p = -0.674$ are achieved [7]. The maximum values of hydrostatic ECF k_h^* at different values of m_p (Fig. 2e) are approximately 1.3–1.5 times larger than the value of the same ECF of PMN–0.33PT SC ($k_h = 0.167$, as follows from experimental data [7]). In general, we note the good agreement between the volume fractions of ECFs calculated by means of the EMM and FEM (Fig. 2).

The prediction of the most extreme values and anisotropy of ECFs (Fig. 2) can aid the design and manufacture of advanced 0–3–0 composites based on PMN–0.33PT and related SCs with high piezoelectric activity. The composite architecture plays an important role in forming the piezoelectric response and ECFs, especially in the presence of the SC component with the large piezoelectric coefficients d_{3j}. The large anisotropy of ECFs of the studied composite is of significance for applications in medical devices with ultrasonic pulse – echo antennae. The large values of hydrostatic ECF are important for hydroacoustic applications. The results discussed in the present paper are of interest for specialists working in the field of modern piezoelectric sensors, transducers and hydrophones.

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