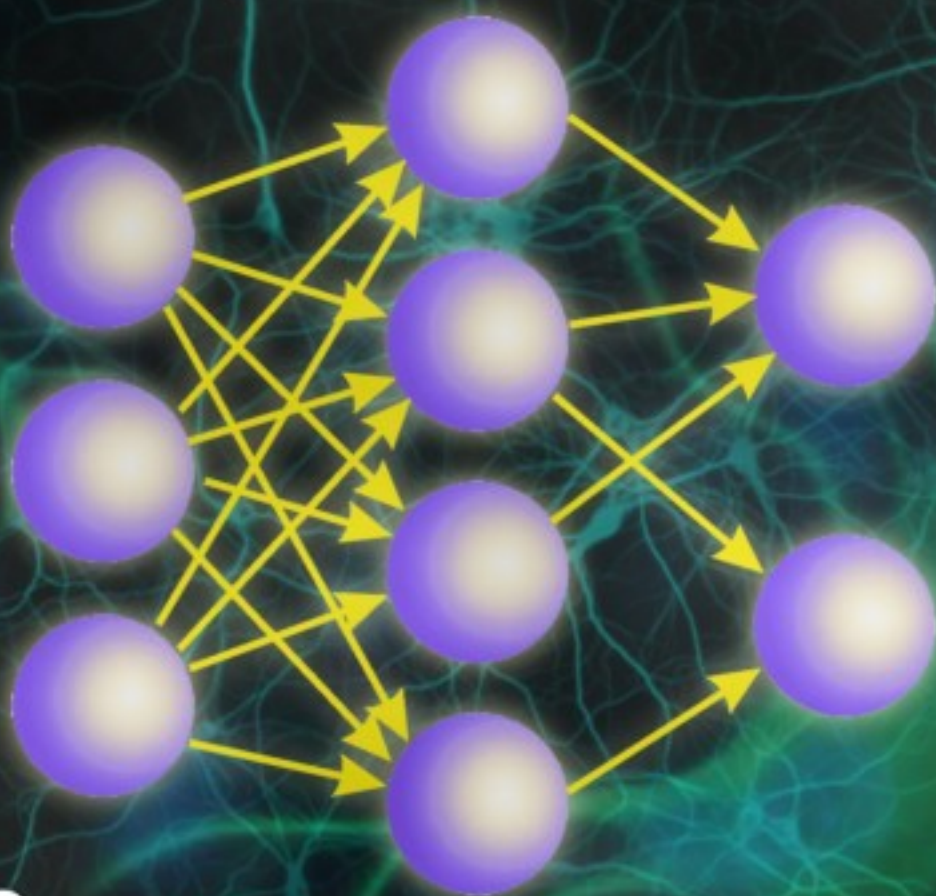


ISSN 1726-5479

SENSORS & **TRANSDUCERS** **10**^{vol. 109}/**09**



Soft Sensors and Artificial Neural Networks

International Frequency Sensor Association Publishing





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Volume 109
Issue 10
October 2009

www.sensorsportal.com

ISSN 1726-5479

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Remarkable Electromechanical Coupling in the 2–2 Composite Based on Single-domain PMN–0.33PT Crystal

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Received: 9 July 2009 / Accepted: 23 October 2009 / Published: 30 October 2009

Abstract: A novel parallel-connected 2–2 single-domain $0.67\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3 - 0.33\text{PbTiO}_3$ crystal / polymer composite with various orientations of polarization vectors of the components is proposed to analyze behavior of electromechanical coupling factors k_{3j}^* and k_h^* where $j = 1, 2$ and 3 . It is shown that the combination of the highly piezo-active relaxor-ferroelectric single-domain component and the piezoelectric polymer provides considerable values of k_h^* ($\min k_h^* \approx -0.8$ and $\max k_h^* \approx 0.7$) and $|k_{33}^*|$ (about 0.9). The active role of the polarization orientation effect and the composite structure in attaining the high performance is emphasized in this work. A strong correlation between k_h^* and the hydrostatic piezoelectric coefficient is first revealed near $\min k_h^*$ and $\max k_h^*$ of the 2–2 composite. Some advantages concerned with the presence of the single-domain component in the 2–2 composite are discussed in connection with the large values of k_{3j}^* and k_h^* as well as with the considerable anisotropy of k_{3j}^* . Copyright © 2009 IFSA.

Keywords: 2–2 composite, Electromechanical coupling factor, Piezoelectric coefficients, Polarization orientation effect

1. Introduction

Outstanding electromechanical properties of perovskite-type relaxor-ferroelectric solid solutions of $(1-x)\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3 - x\text{PbTiO}_3$ (PMN- x PT) and $(1-x)\text{Pb}(\text{Zn}_{1/3}\text{Nb}_{2/3})\text{O}_3 - x\text{PbTiO}_3$ (PZN- x PT) are shown in single crystals (SCs) with engineered domain structures and compositions near the morphotropic phase boundary at room temperature [1–4]. In the last decade these SCs are used to manufacture advanced piezo-active composites, for example, PZN-0.08PT / polymer [5], PMN-0.33PT / epoxy [6] and PMN-0.30PT / epoxy [7] with 1–3 connectivity. The high performance of these and related composites is important for modern piezotechnical applications, including active elements of modern sensors, actuators, hydrophones, and other devices. As is known, changes in domain structures or a poling direction of the aforementioned SCs lead to appreciable changes in their electromechanical properties [1, 3, 4, 8, 9]. At the moment PMN-0.33PT is the only relaxor-ferroelectric SC for which full sets of the electromechanical constants have been measured in both the single-domain (3 m symmetry) [3, 4] and polydomain (or domain engineered, 4 mm symmetry) [1] states at room temperature. The piezoelectric properties of the single-domain PMN-0.33PT SC were analyzed for various orientations of its main crystallographic axes [4, 8].

In earlier studies no attempt has been made to estimate parameters of novel composites based on the single-domain PMN-0.33PT SC. However, according to experimental room-temperature data [4], this material is of interest due to a combination of the large piezoelectric coefficients $d_{15}^{(1)}$ and $d_{22}^{(1)}$ and the moderate piezoelectric coefficients $d_{31}^{(1)}$ and $d_{33}^{(1)}$ (Table 1). This circumstance stimulates a study on a performance of the composite wherein the single-domain PMN-0.33PT SC could show various electromechanical properties owing to changes on the orientation of the crystallographic axes. To the best of our knowledge, electromechanical coupling factors of the composites based on relaxor-ferroelectric SCs were earlier considered fragmentarily and for structures of the 1–3 type only [11, 12]. The aim of the present paper is to study features of electromechanical coupling of the parallel-connected 2–2 single-domain PMN-0.33PT / polymer composite on taking into account various poling orientations of its components.

2. Model of the Composite and its Effective Electromechanical Properties

We consider the 2–2 parallel-connected SC / polymer composite with a regular distribution of layers on the OX_1 direction (Fig. 1). The SC and polymer layers are assumed to be lengthy on the OX_2 and OX_3 directions. An orientation of the spontaneous polarization vector $\mathbf{P}_s^{(1)}$ of each SC layer is characterized by an angle θ (Fig. 1, inset 1). In this work a rotation of the $\mathbf{P}_s^{(1)}$ vector is taken into account in the (X_2OX_3) plane that is parallel to the interfaces between the layers of the 2–2-composite. The polymer layers of this composite can be either ferroelectric (poled along a certain direction) or showing no ferroelectric and piezoelectric properties. It is assumed that the remanent polarization vector $\mathbf{P}_r^{(2)}$ of ferroelectric polymer over the whole composite sample is characterized by one of the following orientations: $\mathbf{P}_r^{(2)} \uparrow\uparrow OX_3$, $\mathbf{P}_r^{(2)} \uparrow\uparrow OX_2$ or $\mathbf{P}_r^{(2)} \uparrow\downarrow OX_3$ (Fig. 1, insets 2–4). A manufacturing of the 2–2 SC / polymer composite with various orientations of the $\mathbf{P}_s^{(1)}$ and $\mathbf{P}_r^{(2)}$ vectors can be succeeded by a consequent poling of the components. In this work the composite comprising PMN-0.33PT and polyvinylidene fluoride (PVDF) is studied. As is known from experimental data, coercive fields $E_c^{(n)}$ of the single-domain PMN-0.33PT SC ($n = 1$) and PVDF ($n = 2$) obey condition [13, 14] $E_c^{(1)} \ll E_c^{(2)}$, that favours the consequent poling of the components chosen.

Table 1. Room-temperature elastic compliances $s_{ab}^{(n),E}$ (in 10^{-12} Pa $^{-1}$), piezoelectric coefficients $d_{ij}^{(n)}$ (in pC N $^{-1}$) and relative dielectric permittivity $\varepsilon_{pp}^{(n),\sigma} / \varepsilon_0$ of the single-domain PMN–0.33PT SC [4] and poled PVDF [10].

Components	$s_{11}^{(n),E}$	$s_{12}^{(n),E}$	$s_{13}^{(n),E}$	$s_{14}^{(n),E}$	$s_{33}^{(n),E}$	$s_{44}^{(n),E}$	$s_{66}^{(n),E}$
PMN–0.33PT SC	62.2	–53.8	–5.6	–166.2	13.3	511.0	232.0
PVDF with $P_r^{(2)} \uparrow \uparrow OX_3$	333	–148	–87.5	0	299	$1.90 \cdot 10^4$	943
Components	$d_{15}^{(n)}$	$d_{22}^{(n)}$	$d_{31}^{(n)}$	$d_{33}^{(n)}$	$\varepsilon_{11}^{(n),\sigma} / \varepsilon_0$	$\varepsilon_{33}^{(n),\sigma} / \varepsilon_0$	
PMN–0.33PT SC	4100	1340	–90	190	3950	640	
PVDF with $P_r^{(2)} \uparrow \uparrow OX_3$	–38	0	10.42	–33.64	7.513	8.431	

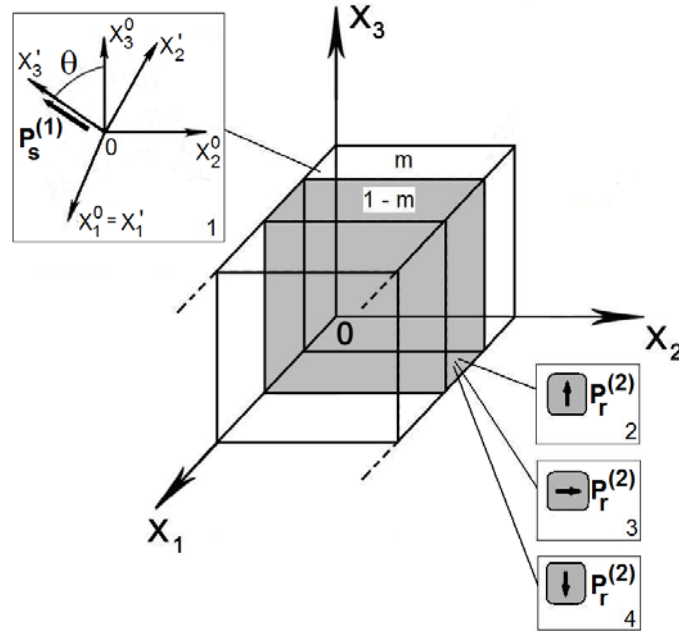


Fig. 1. Schematic of the 2–2 SC / polymer composite with parallel-connected layers. $(X_1X_2X_3)$ is the rectangular co-ordinate system, m and $1 - m$ are volume fractions of SC and polymer, respectively, $P_s^{(1)}$ and $P_r^{(2)}$ are the spontaneous polarization vector of SC and the remanent polarization vector of polymer, respectively. In inset 1 θ is the angle that characterizes the rotation of the main crystallographic axes $(X_1^o X_2^o X_3^o) \rightarrow (X_1' X_2' X_3')$ and the orientation of the $P_s^{(1)}$ vector in each single-domain SC layer. Orientations of the remanent polarization vector of polymer $P_r^{(2)} \uparrow \uparrow OX_3$, $P_r^{(2)} \uparrow \uparrow OX_2$ and $P_r^{(2)} \uparrow \downarrow OX_3$ are shown in insets 2, 3 and 4, respectively.

Effective electromechanical constants and other parameters of the composite are evaluated with use of full sets of elastic compliances $\|s^{(n),E}\|$ (measured at electric field $E = \text{constant}$), piezoelectric coefficients $\|d^{(n)}\|$ and dielectric permittivities $\|\varepsilon^{(n),\sigma}\|$ (measured at mechanical stress $\sigma = \text{constant}$) of components (Table 1). We describe the rotation of the main crystallographic axes of SC $(X_1^o X_2^o X_3^o) \rightarrow (X_1' X_2' X_3')$ (Fig. 1, inset 1) with the $\|r\|$ matrix (3×3) and determine the full set of electromechanical constants in the $(X_1' X_2' X_3')$ system as follows:

$$s_{quv}^{(1),E} = r_{qa} r_{tb} r_{uc} r_{vd} (s_{abcd}^E)_0, d_{efg}^{(1)} = r_{ej} r_{fk} r_{gl} (d_{jkl})_0 \text{ and } \varepsilon_{mp}^{(1),\sigma} = r_{mh} r_{pi} (\varepsilon_{hi}^\sigma)_0, \quad (1)$$

where $(s_{abcd}^E)_0$, $(d_{jkl})_0$ and $(\varepsilon_{hi}^\sigma)_0$ are electromechanical constants related to the $(X_1^o X_2^o X_3^o)$ system, $OX_w^o \parallel OX_w$ at $\theta = 0^\circ$, $w = 1, 2$ and 3 , and non-zero elements of the $\| r \|$ matrix are $r_{11} = 1$, $r_{22} = r_{33} = \cos \theta$, and $r_{23} = -r_{32} = \sin \theta$. Hereafter we use the two-index form [15] of electromechanical constants from Eqs. (1) and consider effective parameters with respect to the $(X_1 X_2 X_3)$ system of the composite sample (Fig. 1). The rotation angle θ is varied from 0° to 180° . The electromechanical properties of the n th component of the composite in the $(X_1 X_2 X_3)$ system are described using the Van Dyke matrix [15]

$$\| C^{(n)} \| = \begin{pmatrix} \| s^{(n),E} \| & \| d^{(n)} \|^T \\ \| d^{(n)} \| & \| \varepsilon^{(n),\sigma} \| \end{pmatrix} \quad (2)$$

where $n = 1$ and 2 , and superscript T denotes the transposed matrix.

A procedure for averaging the electromechanical properties from Eq. (2) is realized along the OX_1 axis by taking into consideration nine boundary conditions [16] for electric and mechanical fields in the adjacent layers of the composite sample (Fig. 1). These boundary conditions at $x_1 = \text{constant}$ involve the continuity of three normal components of the mechanical stress (i.e., σ_{11} , σ_{12} and σ_{13}), three tangential components of the mechanical strain (i.e., ξ_{22} , ξ_{23} and ξ_{33}), one normal component of the electric displacement (i.e., D_1), and two tangential components of the electric field (i.e., E_2 and E_3). Following this procedure for averaging, the effective electromechanical properties of the 2–2 composite are determined from a matrix

$$\| C^* \| = [\| C^{(1)} \| \| M \| m + \| C^{(2)} \| (1 - m)] [\| M \| m + \| I \| (1 - m)]^{-1} \quad (3)$$

that is represented in the form similar to that shown in Eq. (2). In Eq. (3) $\| M \|$ is the 9×9 matrix that describes the aforementioned boundary conditions at $x_1 = \text{constant}$, $\| I \|$ is the identity 9×9 matrix, m is the volume fraction of SC, and $\| C^{(n)} \|$ is taken from Eq. (2). The full set of effective constants of the composite s_{ab}^{*E} , d_{ij}^* and $\varepsilon_{jh}^{*\sigma}$ is determined from matrix (3). These constants are determined in the longwave approximation, i.e., on condition that the wavelength of an external acoustic field is much more than a width of each layer of the composite (Fig. 1).

Based on the averaging procedure, we consider the volume-fraction (m) and orientation (θ) dependences of effective electromechanical coupling factors

$$k_{3j}^* = d_{3j}^* / (\varepsilon_{33}^{*\sigma} s_{jj}^{*E})^{1/2} \quad (4)$$

and hydrostatic electromechanical coupling factor

$$k_h^* = d_h^* / (\varepsilon_{33}^{*\sigma} s_h^{*E})^{1/2}, \quad (5)$$

where $j = 1, 2$ and 3 and the hydrostatic piezoelectric coefficient and elastic compliance of the composite

$$d_h^* = \sum_{i=1}^3 \sum_{j=1}^3 d_{ij}^* \quad \text{and} \quad s_h^{*E} = \sum_{a=1}^3 \sum_{b=1}^3 s_{ab}^{*E} \quad (6)$$

are expressed in terms of effective electromechanical constants d_{ij}^* and s_{ab}^{*E} , respectively.

3. Results and Discussion

3.1. Electromechanical Coupling Factors k_{3j}^*

At first we consider the effective parameters (4) and (5) of the composite that contains PVDF with $P_r^{(2)} \uparrow \downarrow OX_3$ (Fig. 1, inset 4). The polymer component poled in this direction has the piezoelectric coefficients $d_{31}^{(2)} < 0$, $d_{33}^{(2)} > 0$ and $d_{15}^{(2)} > 0$, and their signs coincide with those of SC in the main crystallographic axes (Table 1).

Examples of graphs of $k_{3j}^*(m, \theta)$ are shown in Fig. 2(a) – (c). It is seen that an anisotropy of k_{3j}^* takes place at various m and θ values. In this connection one can single out the following ranges of m and θ :

- (i) at $0.1 < m < 0.6$ and $1^\circ \leq \theta \leq 10^\circ$, inequalities $0 < |k_{31}^*| < 0.1$ and $|k_{31}^*| < |k_{32}^*| < k_{33}^*$ hold, and $k_{31}^*(m, \theta)$ passes the zero value (Fig. 2(a)) due to changes in $\text{sgn} d_{31}^*$ that is caused by the orientation effect in the presence of interfaces $x_1 = \text{const}$ (Fig. 1),
- (ii) at $0.1 < m < 0.8$ and $89^\circ \leq \theta \leq 91^\circ$, inequalities $0 < |k_{32}^*| < 0.1$ and $|k_{32}^*| < k_{31}^* < |k_{33}^*|$ hold that may be concerned with small angles between the spontaneous polarization vector $P_s^{(1)}$ and the OX_2 axis (Fig. 1),
- (iii) at $0.55 < m \leq 0.8$ and $97^\circ \leq \theta \leq 106^\circ$, electromechanical coupling factors obey conditions $|k_{3j}^*| > 0.5$ and $\text{sgn} k_{31}^* = \text{sgn} k_{32}^* = -\text{sgn} k_{33}^*$, and
- (iv) inequalities $k_{33}^* / |k_{31}^*| > 5$ and $k_{33}^* / |k_{32}^*| > 5$ simultaneously hold at $0.14 < m < 0.28$ and $\theta = 177^\circ$, $0.17 < m < 0.52$ and $\theta = 178^\circ$ and $0.29 < m < 0.75$ and $\theta = 179^\circ$.

Such a variety of interrelations between the k_{3j}^* values (Fig. 2(a) – (c)) is concerned with the anisotropy of electromechanical properties of SC at various orientations of the $P_s^{(1)}$ vector. Undoubtedly, the composite with $|k_{3j}^*| > 0.5$ is of interest for transducer and other piezotechnical applications.

3.2. Hydrostatic Electromechanical Coupling Factor k_h^*

In work [12] the volume-fraction dependence of hydrostatic coupling factor k_h^* was first predicted and analyzed for a 1–2–2 composite based on the polydomain PMN–0.33PT SC poled along the [001] perovskite unit-cell direction. The electromechanical interaction between the highly piezo-active SC rods and the laminar polymer matrix favours the considerable k_h^* values in a wide volume-fraction range. For example, in the 1–2–2 polydomain PMN–0.33PT / araldite / elastomer composite $\max k_h^* = 0.473$ attained whereas the SC component has $k_h^{(1)} = 0.167$ only and is surrounded by two piezo-passive polymers [12].

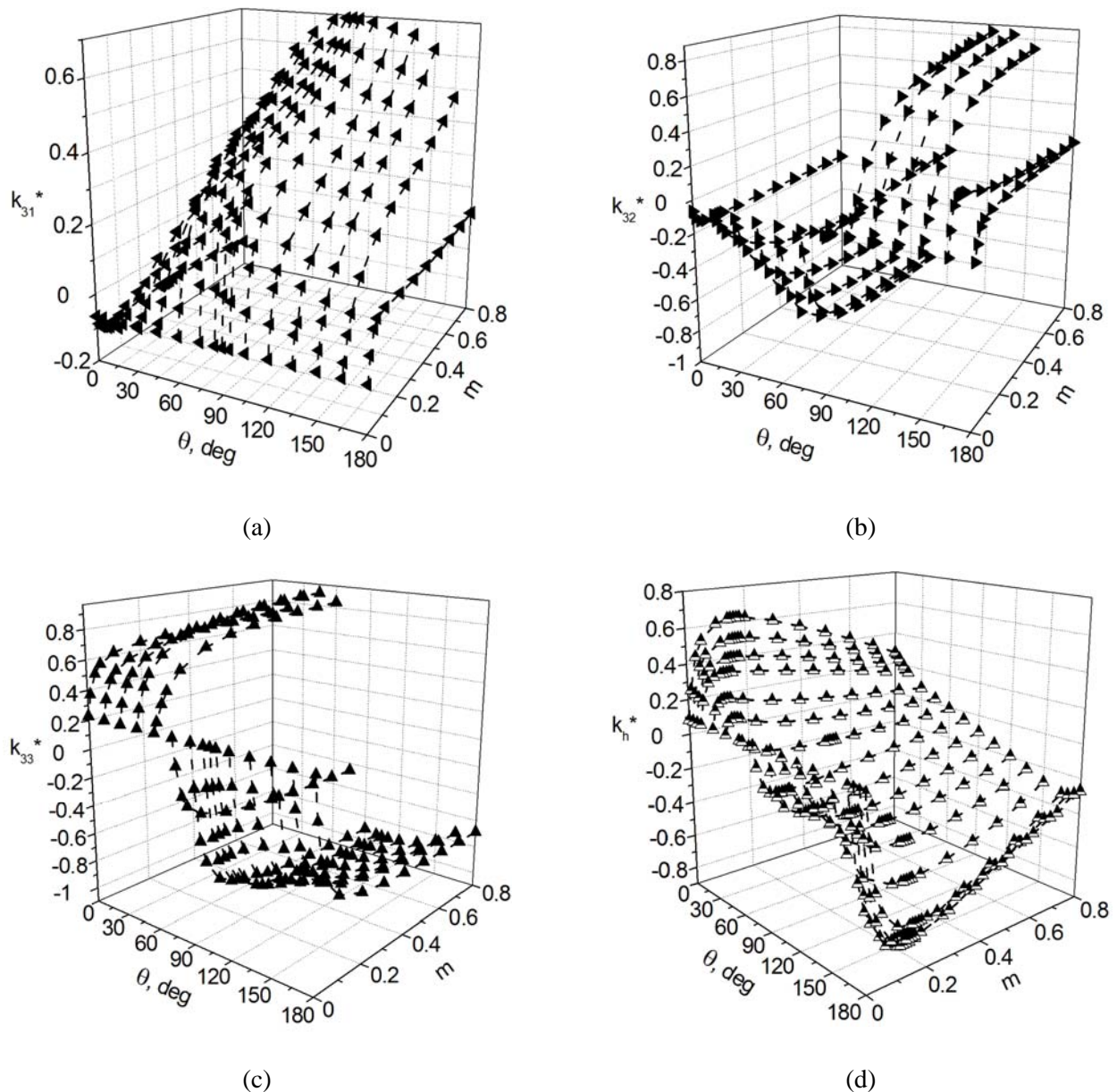


Fig. 2. Volume fraction (m) and orientation (θ) dependences of effective electromechanical coupling factors k_{3j}^* (a–c) and k_h^* (d) of the 2–2 single-domain PMN–0.33PT / PVDF composite with $P_r^{(2)} \uparrow \downarrow OX_3$.

The present results show that the 2–2 composite based on the single-domain PMN–0.33PT SC with $k_h^{(1)} = 0.048$ is characterized by a stronger hydrostatic electromechanical coupling (Fig. 2(d) and Table 2) when the polymer component is piezoelectric. The piezoelectric polymer component (i.e., PVDF) influences the $k_h^*(m, \theta)$ dependence so that $|\min k_h^*| \neq \max k_h^*$ (Table 2) and the $P_s^{(1)}$ vectors related to these extreme points are not parallel. Small differences between the extreme values found for the related composites with different orientations of $P_r^{(2)}$ in the PVDF layers are accounted for by the volume fractions $m > 0.1$: in this range the piezoelectric properties of SC component play the leading role in forming the hydrostatic piezoelectric coefficient d_h^* (see Eqs. (5) and (6)) and, therefore, the hydrostatic electromechanical coupling of the composite.

Table 2. Minimum and maximum values of hydrostatic electromechanical coupling factor $k_h^*(m, \theta)$ calculated for the 2–2 single-domain PMN–0.33PT / polymer composite.

Polymer	$\min k_h^*(m, \theta)$	$\max k_h^*(m, \theta)$
PVDF with $\mathbf{P}_r^{(2)} \uparrow \downarrow OX_3$	-0.792 ($m = 0.16$ and $\theta = 169^\circ$)	0.626 ($m = 0.16$ and $\theta = 0^\circ$)
PVDF with $\mathbf{P}_r^{(2)} \uparrow \uparrow OX_3$	-0.782 ($m = 0.16$ and $\theta = 169^\circ$)	0.639 ($m = 0.16$ and $\theta = 0^\circ$)
PVDF with $\mathbf{P}_r^{(2)} \uparrow \uparrow OX_2$	-0.766 ($m = 0.16$ and $\theta = 172^\circ$)	0.667 ($m = 0.20$ and $\theta = 0^\circ$)
Araldite ^a	-0.339 ($m = 0.47$ and $\theta = 180^\circ$)	0.339 ($m = 0.47$ and $\theta = 0^\circ$)
Polyurethane ^b	-0.337 ($m = 0.34$ and $\theta = 180^\circ$)	0.337 ($m = 0.34$ and $\theta = 0^\circ$)
Elastomer ^c	-0.218 ($m = 0.07$ and $\theta = 180^\circ$)	0.218 ($m = 0.07$ and $\theta = 0^\circ$)

^a Experimental values of elastic and dielectric constants were taken from work [17]

^b Experimental values of elastic and dielectric constants were taken from work [18]

^c Experimental values of elastic and dielectric constants were taken from work [19]

A comparison of data from Table 2 suggests that the poled polymer plays the important role of a link providing the appreciable electromechanical interaction between the SC layers of the composite (Table 1) and the considerable piezoelectric response of the composite as a whole. Surprisingly, the related composites comprising one of the piezo-passive polymer components (e.g., polyurethane with smaller $\varepsilon_{pp}^{(2)}$ [18] or elastomer with larger $|s_{ab}^{(2)}|$ [19]) show the smaller values of k_h^* (Table 2) in comparison with the single-domain PMN–0.33PT / PVDF composite. We also add for comparison that a 1–3 PZT ceramic / porous polyurethane composite [18] would have k_h^* from about 0.11 to 0.57 on dependence of the elastic properties of the porous matrix. In this 1–3 composite a ratio of $k_h^* / k_h^{(1)}$ is predicted to be varied from 1.4 to 7.3.

A very important feature of the $k_h^*(m, \theta)$ dependence of the single-domain PMN–0.33PT / PVDF composite consists in a strong correlation between k_h^* and d_h^* (Fig. 3). It means that a product $\varepsilon_{33}^{*\sigma} s_h^{*E}$ from Eq. (5) plays a passive role in forming the $k_h^*(m, \theta)$ dependence at $m > 0.1$. Such behaviour may be concerned with a compensation of increasing $\varepsilon_{33}^{*\sigma}$ with decreasing s_h^{*E} of the studied composite, and this effect can be of an independent interest. It should be added that, in the vicinity of the extreme points of $k_h^*(m, \theta)$, the hydrostatic piezoelectric coefficient d_h^* attains 300 – 400 pC / N (Fig. 3) and is of value for hydrophone applications.

4. Conclusions

The present paper is concerned with the study on electromechanical coupling factors k_{3j}^* and k_h^* of the 2–2 composite based on the single-domain PMN–0.33PT SC. The volume-fraction and orientation behavior of electromechanical coupling is analyzed for the low-symmetry composite wherein the poling directions of the SC and polymer components can be rotated in the plane of interfaces separating these components. The results point out opportunities associated with use of the single-domain SC component and with preferable orientations of its spontaneous polarization vector $\mathbf{P}_s^{(1)}$. The present study shows the important role of the $\mathbf{P}_s^{(1)}$ orientation in forming the large values of $|k_{3j}^*|$ and $|k_h^*|$ in the wide volume-fraction range. A comparison of data on the composites with different polymers suggests that poled PVDF promotes the largest values of $|k_{3j}^*|$ and $|k_h^*|$ due to the effective

electromechanical interaction between two piezo-active components. A ratio of $k_h^*(m, \theta) / k_h^{(1)}$ calculated for the single-domain PMN–0.33PT / PVDF composites with the certain $P_r^{(2)}$ orientations is varied from -16.5 to 13 , and such a range has no analogues among the piezo-active composites studied earlier.

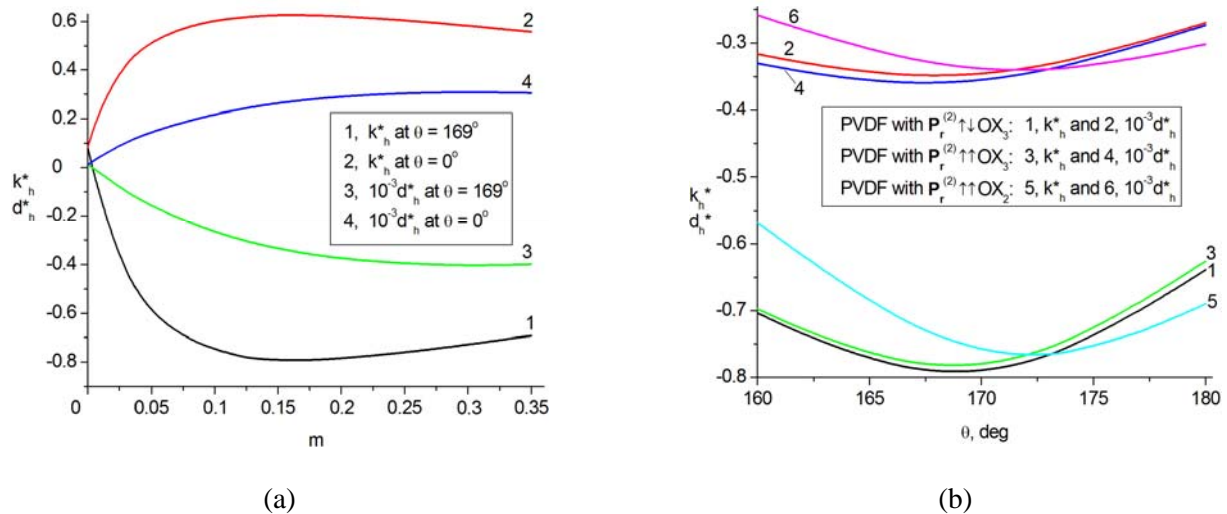


Fig. 3. Correlation between hydrostatic parameters of composites near absolute min $k_h^*(m, \theta)$ and max $k_h^*(m, \theta)$.

Graph (a) has been built for the 2–2 single-domain PMN–0.33PT / PVDF composite with $P_r^{(2)} \uparrow \downarrow OX_3$ at $m = 0.16$. In graphs (a) and (b) the hydrostatic piezoelectric coefficient d_h^* in pC / N.

The presence of the piezo-passive polymer component, being even more compliant or having a lower dielectric permittivity in comparison with PVDF, results in appreciable decreasing $|k_h^*|$. It should be added that in earlier papers no electromechanical coupling factors of the SC / polymer composite were analyzed on varying both the volume fraction m and the rotation angle θ . The results of our study can be of interest for transducer and hydroacoustic applications of the advanced 2–2 composites based on relaxor-ferroelectric SCs. One can believe that high performance predicted and discussed in the present paper can stimulate the manufacturing of novel piezo-active composites with both considerable electromechanical coupling factors and high piezoelectric activity.

Acknowledgements

The authors would like to thank Prof. Dr. R. Stevens (University of Bath, UK), Prof. Dr. P. Bisegna (University of Rome “Tor Vergata”, Italy), Prof. Dr. I.A. Parinov (Southern Federal University, Russia), and Dr. M. Kamlah (Karlsruhe Research Centre, Germany) for their interest in the research problems. This work was partially supported by the administration of the Southern Federal University (Project No. 11.1.09f on basic research), and this support is gratefully acknowledged.

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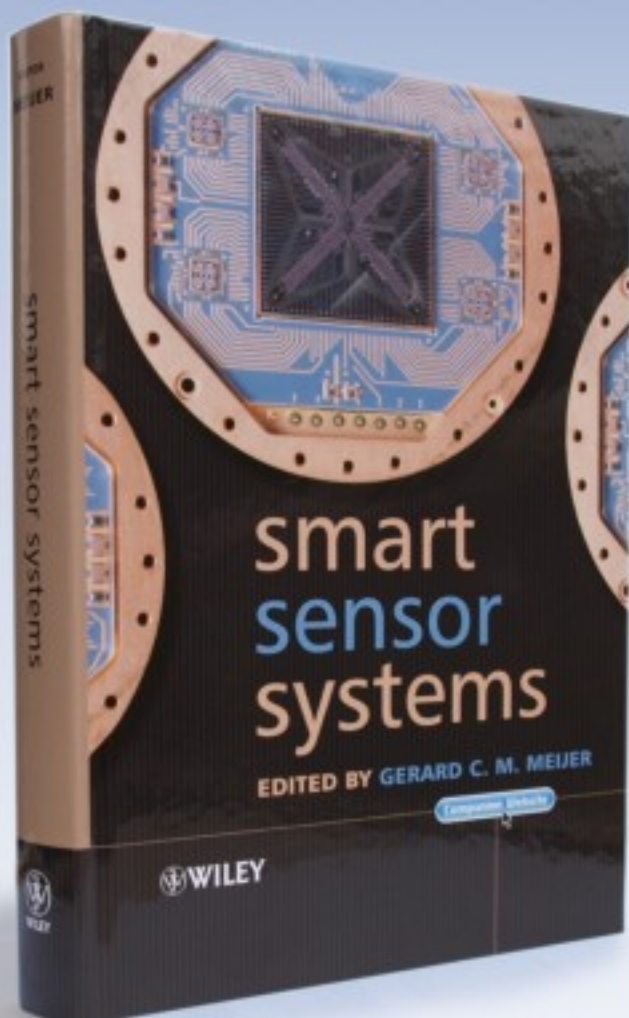
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