EFFECTIVE PROPERTIES OF A TRANSVERSELY ISOTROPIC PIEZOELECTRIC COMPOSITE WITH CYLINDRICAL INCLUSIONS

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UDC 539.31.001:678.067:537

The effective properties of heterogeneous media are determined mainly within the framework of statistical and model approaches. The generality of the statistical theories in these cases is usually due either to the isometric form of the components or to the determination of correlation functions. The correctness of statistical approaches is checked by comparison with the results of calculation of the effective properties on model with prescribed geometric characteristics.

The models of piezoelectric composite materials (PCM) proposed in the literature [1, 2], based on serial and parallel connection schemes and their combinations, contain several simplifying assumptions which make it impossible to correctly account for the interaction of the elastic and electrical subsystems in a nonuniform anisotropic piezotexture.

The authors of [3] calculated the effective properties of a laminated PCM consisting of alternating symmetrical piezoelectric layers 4 mm thick (the axes of symmetry of the components were parallel and were located in the planes of the interfaces). The use of this method to perform calculations for a two-phase PCM with its symmetry axis perpendicular to the plane of the interface between the layers for the most part substantiated the statistical theory in [4] (only λ^*_1 , λ^*_3 , χ^*_1 , e^*_{31} did not coincide with the exact results obtained in [3]). Making the requirements in regard to the accuracy of the prediction of PCM effective properties more stringent is of more than just academic interest. The increasing range of components being used in the development of PCM's (the use of polymers and ferroelectric materials, their polarization, an increase in the surface per unit volume of the PCM, the effect of the adhesive properties of the polymers) and the attendant manifestation of new effects require accurate determination of the effective properties of PCM's - albeit within the framework of the linear theory of electroelasticity.

1. A PCM reinforced with unidirectional fibers is usually produced by placing ferroelectric ceramic rods in a polymer matrix or by filling holes drilled in a ceramic block with a polymer and subsequently polarizing the specimen. The symmetry of such a PCM is ∞m .

We will assume that the distribution of the fibers in the plane of isotropy is random. We surround all of the fibers with as many nonintersecting cylindrical surfaces as possible and we assume that each fiber (r = a) and its surrounding cylindrical matrix (r = b) - acompound cylinder - are located in a nonuniform medium having effective properties. Since the PCM is assumed to be macroscopically uniform, the average mechanical (σ^*, ξ^*) and electrical (E^*, D^*) fields in the specimen will be constant and equal to the corresponding quantities at the boundary of the compound cylinder (r = b). This approximation, usually used in the self-consistent field method, makes it possible to employ the hypothesis of equivalent homogeneity and to calculate the effective properties of a PCM on the basis of a single compound cylinder [5]. In this sense, the formulas obtained should be considered approximate in regard to actual PCM's and should most closely reflect the properties of PCM's having fibers of different cross sections with a/b = const. The results presented here will be exact for PCM's which can be represented as a polydisperse model of a medium with cylindrical inclusions [5, 6].

We place the origin of the cylindrical coordinate system at the center of the coaxial cylinders. The z axis is directed along the symmetry axis. With allowance for the symmetry of the components ∞m, the equations of state of the piezoelectric medium [7] will have the form

$$\xi_{z} = s_{13}{}^{E}\sigma_{r} + s_{13}{}^{E}\sigma_{\theta} + s_{33}{}^{E}\sigma_{z} + d_{33}E_{z}, \qquad \xi_{r} = s_{11}{}^{E}\sigma_{r} + s_{12}{}^{E}\sigma_{\theta} + s_{13}{}^{E}\sigma_{z} + d_{31}E_{z}; \tag{1}$$

Rostov-on-Don State Pedagogical Institute. Translated from Mekhanika Kompozitnykh Materialov, No. 1, pp. 62-69, January-February, 1989. Original article submitted May 27, 1988.

$$\xi_{\theta} = s_{12}{}^{E}\sigma_{r} + s_{11}{}^{E}\sigma_{\theta} + s_{13}{}^{E}\sigma_{z} + d_{31}E_{z}; \qquad D_{z} = d_{31}\sigma_{r} + d_{31}\sigma_{\theta} + d_{33}\sigma_{z} + \varepsilon_{33}\sigma_{z}E_{z}; D_{r} = d_{15}\sigma_{zr} + \varepsilon_{11}{}^{\sigma}E_{r}; \qquad 2\xi_{zr} = s_{55}{}^{E}\sigma_{zr} + d_{15}E_{r}; \qquad D_{\theta} = d_{15}\sigma_{z\theta} + \varepsilon_{11}{}^{\sigma}E_{\theta};$$
(2)
$$2\xi_{z\theta} = s_{55}{}^{E}\sigma_{z\theta} + d_{15}E_{\theta};$$
(3)

$$2\xi_{r\theta} = s_{66}\sigma_{r\theta}.$$

The idea behind the method in [3], first proposed in [8], is that, in the case of unidimensional inhomogeneity, certain components of the electric and elastic fields can be determined from the system of differential equations which describes the thermodynamically equilibrium distribution of these fields in a nonuniform medium [9-11]:

div
$$\mathbf{D} = 0$$
; rot $\mathbf{E} = 0$; Div $\boldsymbol{\sigma} = 0$; Ink $\boldsymbol{\xi} = 0$. (4)

Then the remaining components of the average fields are determined through the equations of state (found by simple integration) with allowance for the coordinate dependence of the material tensors. This approach ensures the solution of the averaging problem.

In materials reinforced with fibers, the ratio of the length to the diameter of the fibers is usually very large. Thus, all of the quantities in (1-4) are independent of z ($\partial/\partial z = 0$).

We will determine the effective constants of a PCM by solving boundary-value problems of the theory of electroelasticity for a compound cylinder. Here, we use the averaging method [3]. For more complex cases, we obtain a solution on the basis of variational estimates, comparing the results for different types of boundary conditions (Foigt-Reiss scheme). Two basic cases are possible in the measurement of the physicomechanical properties of a PCM.

2. In the first case, external loading does not alter the symmetry of the medium. If an electric field E_z^* is applied in the PCM along the Z axis, then the symmetry of the problem remains ∞m , and all of the quantities describing the state of the medium will depend only on r. Then it follows from the condition rot E = 0 that E_z is independent of r and, thus, is equal to its mean value $E_z = \langle E_z \rangle (\langle E_z \rangle = E_z^*)$. One of the strain compatibility conditions (4), meanwhile, implies that ξ_z is independent of r, i.e., $\xi_z = \langle \xi_z \rangle$. The equation of equilibrium $d\sigma_r/dr + (\sigma_r - \sigma_\theta)/r = 0$ needed in this case is satisfied identically if we take

$$\sigma_r = F/r; \quad \sigma_\theta = dF/dr. \tag{5}$$

We find the stress function F(r) from the strain compatibility condition $d\xi_{\theta}/dr + (\xi_{\theta} - \xi_r)/r = 0$, having inserted the strain components from (1) with allowance for (5). Then for F we have the equation $d^2F/dr^2 + (dF/dr)/r - F/r^2 = 0$, the general solution of which is

$$F = C \cdot r + D/r. \tag{6}$$

We find the constants C and D from the boundary conditions on the surfaces of the cylinders for

$$\mathbf{r} = \mathbf{a} \quad \mathbf{\sigma}'_{\mathbf{r}} = \mathbf{\sigma}''_{\mathbf{r}}, \quad \mathbf{U}'_{\mathbf{r}} = \mathbf{U}''_{\mathbf{r}}; \quad \text{for} \quad \mathbf{r} = \mathbf{b} \quad \mathbf{\sigma}''_{\mathbf{r}} = \mathbf{0}. \tag{7}$$

Here and below, one prime is used to denote quantities pertaining to the internal cylinder $(0 \le r \le a)$, while two primes denote quantities pertaining to the external cylinder ($a \le r \le b$).

To determine the strain $\langle \xi_Z \rangle$, we use the integral condition for a mechanically free compound cylinder:

$$\langle \sigma_z \rangle = \int_0^{2\pi} d\theta \int_0^b \sigma_z(r) r dr / \pi b^2 = 0.$$
(8)

Equation (8) is an example of determination of the mean value of electric and mechanical fields. These mean values, prescribed in the composite in accordance with the external conditions, will henceforth be denoted with an asterisk.

Having combined (5-8), we find the distribution of the stresses, strains, and electric displacement for system (1) in relation to r. Then, on the basis of the hypothesis of equivalent homogeneity [5], we can find the effective permittivity of a mechanically free specimen and the effective piezomoduli (in the reciprocal piezoeffect) from the definitions

$$\varepsilon^{*\sigma_{33}} = \langle D_z \rangle / E^*_{z}; \ d^*_{33} = \langle \xi_z \rangle / E^*_{z}; \ d^*_{31} = \langle (\xi_r + \xi_\theta) / 2 \rangle / E^*_{z}; \ d^*_{v} = \langle \operatorname{Sp} \xi \rangle / E^*_{z}$$

The results of the calculations have the form

$$\varepsilon_{33}^{*\sigma} = \langle \varepsilon_{33}^{\sigma} \rangle - m_2 m_2 (\tilde{a}_{33}^2 s_c + 2 \tilde{a}_{31}^2 [s_{33}^E] - 4 \tilde{a}_{31} \tilde{a}_{33} [s_{13}^E]) / A_c;$$
(9)

$$d^*_{33} = (s_c \cdot \langle d_{33}s_{33}^E] - 2[s_{13}^E] \cdot \langle d_{33}s_{13}^E] + 2m_1 m_2 d_{31} s_6) / A_c;$$

$$d^*_V = d^*_{33} + 2d^*_{31}; (10)$$

 $d^*_{31} = \left[\left(2m_1 d'_{31} s_{11}''^E + m_2 d''_{31} s \right) \cdot \left[s_{33}^E \right] - 2\left[s_{13}^E \right] \cdot \langle d_{31} s_{13}^E \right] + m_1 m_2 \tilde{d}_{33} \left(s_{13}''^E - 2s_{11}''^E s_{13}'^E \right) \right] / A_c.$ (11)

Here, $m_1 = a^2/b^2$ and $m_2 = 1 - m_1$ is the volume concentration of the inclusions and the matrix, respectively. The system of notation used below for the other quantities has the form

$$\langle \epsilon_{ij} \circ \rangle = m_1 \epsilon_{ij}'^{\sigma} + m_2 \epsilon_{ij}''^{\sigma}; \ [s_{\alpha\beta}{}^E] = m_1 s_{\alpha\beta}''^E + m_2 s_{\alpha\beta'}{}^E; \ \bar{d}_{i\alpha} = d'_{i\alpha} - d''_{i\alpha}; \langle d_{i\alpha} s_{\beta\gamma}{}^E] = m_1 d'_{i\alpha} s_{\beta\gamma}''^E + m_2 d''_{i\alpha} s_{\beta\gamma'}{}^E; \ s_{\pm} = s_{11}{}^E \pm s_{12}{}^E; \ s_c = [s_{\pm}] + s''_{-}; A_c = s_c [s_{33}{}^E] - 2[s_{13}{}^E]^2; \ s = s'_{\pm} + s''_{-}; \ s_6 = s_{33}{}^{'E} s_{13}{}^{'E} - s_{33}{}^{''E} s_{13}{}^{'E}.$$

The effective piezomoduli for the direct piezoeffect is determined for short-circuit (SC) conditions: $E_z = E_z^* = 0$. To determine the piezomodulus d_{31}^* , it is necessary to apply a plane stress $\sigma_{\perp}^* = \sigma_{\Gamma}^*(b) = \sigma_{\theta}^*(b)$ to the specimen. To determine the piezomodulus d_{33}^* , condition (8) should be replaced by $\langle \sigma_z \rangle = \sigma_z^*$. The bulk piezomodulus is determined for conditions of hydrostatic compression: $\langle \sigma_z \rangle = \sigma_{\Gamma}^*(b) = \sigma_{\theta}^*(b) = -p^*$. Taking these boundary conditions into consideration along with Eqs. (1), (5), and (6) and using the operation of averaging

$$d^*_{31} = \langle D_z \rangle / 2\sigma^*_{\perp} |_{\langle \sigma_z \rangle = 0}; \quad d^*_{33} = \langle D_z \rangle / \sigma^*_z |_{\sigma''_{(b)} = 0}; \quad d^*_V = \langle D_z \rangle / (-p^*)$$

we obtain expressions for the effective piezomoduli which coincide with the expressions found by the previous method. This agreement is due to the fact that, thanks to the high degree of symmetry of the medium, the resulting solutions of the boundary-value problems of electroelasticity theory actually reduce the integrand functions to piecewise-uniform functions. This fact also accounts for the exact solution for $\varepsilon_{33}^{*\sigma}$.

In assigning the boundary conditions for the displacements (strains), it is necessary to examine another system of equations of state in the variables ξ , E:

$$\sigma_{\alpha} = c_{\alpha\beta} E_{\beta} - e_{k\alpha} E_{k}; \quad D_{m} = e_{m\beta} \xi_{\beta} + \varepsilon_{mk} \xi_{k}. \tag{12}$$

Taking the same approach as with (1) and examining the first four equations of this system, we obtain the following solution to the equilibrium equation in displacements (with allowance for the fact that $\xi_z = \langle \xi_z \rangle$): $U_r = Ar + B/r$; $U_z = \langle \xi_z \rangle z$. Then assigning the boundary conditions for the displacements in terms of the strains ($\xi_r = dU_r/dr$; $\xi_\theta = U_r/r$; $\xi_z = \langle \xi_z \rangle$) consistent with the symmetry of the problem, we find the effective piezoconstants $e^*_{k\alpha}$ under SC conditions. To determine e^*_{31} , we create plane strain in the plane of isotropy $\xi_\perp^* = \xi_{r}^{"}(b) = \xi_{\theta}^{"}(b)$. To determine e^*_{33} , we assign $\xi_z^* = \langle \xi_z \rangle$. With allowance for the continuity of the displacements and the stresses at r = a (7), this proves to be sufficient to solve the averaging problem by using the physical definition of the piezoconstants [7]:

$$e^{*}_{31} = \langle D_z \rangle / \langle \xi_r + \xi_0 \rangle |_{\langle \xi_z \rangle = 0}; \quad e^{*}_{33} = \langle D_z \rangle / \xi^*_z |_{\langle \xi_r + \xi_0 \rangle = 0}$$

Then, taking into account the conditions of measurement of the effective piezoconstants, we obtain

$$e^{*}_{31} = \left[2m_{1}e'_{31}c_{11}''^{E} + m_{2}e''_{31}\left(c_{11}'^{E} + c_{12}'^{E} + c_{11}''^{E} - c_{12}''^{E}\right)\right]/c;$$
(13)

$$e^{*}_{33} = \langle e_{33} \rangle - 2m_{1}m_{2}(e^{*}_{31} - e^{*}_{31})(c_{13}^{*E} - c_{13}^{*E})/c;$$

$$c = c_{11}^{*E} - c_{12}^{*E} + [(c_{11}^{E} + c_{12}^{E})].$$
(14)

The effective elastic constants in the first four equations of system (12) are more easily determined than the same constants in the first four equations of system (1), since the elastic moduli $c_{\alpha\beta}^E$ usually are among the elastic properties of the material determined experimentally. When the elastic moduli are measured in the SC regime, $E_z = 0$, and the problem reduces to a purely elastic problem [5, 6]. We will write out the results in the form normally used for piezoelectric materials:

$$c_{11}^{*E} + c_{12}^{*E} = \left[\left(c_{11}^{''E} - c_{12}^{''E} \right) \left\langle c_{11}^{E} + c_{12}^{E} \right\rangle + \left(c_{11}^{''E} + c_{12}^{''E} \right) \left(c_{11}^{''E} + c_{12}^{''E} \right) \right] / c; \tag{15}$$

$$c_{13}^{*E} = \left[2m_1c_{13}'^{E}c_{11}''^{E} + m_2c_{13}''^{E}(c_{11}'^{E} + c_{12}'^{E} + c_{11}''^{E} - c_{12}''^{E})\right]/c;$$
(16)

$$c_{33}^{*E} = \langle c_{33}^{E} \rangle - 2m_1 m_2 (c_{13}'^{E} - c_{13}''^{E})^2 / c.$$
(17)

The variational estimates in [5, 6] show that solutions (15)-(17) are exact for the PCM model being considered. Here, we also satisfy the familiar relations [7] for the effective piezoconstants (10), (11) and (13), (14): $e_{k\alpha}^* = d_{k\beta}^* c_{\beta\alpha}^{*E}$. This confirms the mutual self-consistency of the piezoelectric and elastic problems in the proposed PCM model.

3. In the second case, external loading diminishes the symmetry of the medium. This situation is realized in the measurement of the shear modulus, piezomodulus d_{15}^* , and permittivity $\varepsilon_{11}^{*\sigma}$ (Eqs. (2) and (3)).

To measure $\varepsilon_{11}^{*\sigma}$, we direct the electrical displacement D^{*} (or field) perpendicular to the axis of the cylinders along the X axis. The mechanical state of the medium corresponds to the state of simple shear in the XZ plane [6]. Then the symmetry of the medium + field system is reduced, and all of the quantities will depend on r and θ . The equations of electrical and elastic equilibrium (4) in this case yield a system of two second-order differential equations

$$\nabla^{2}(e_{15}U_{z}-e_{11}\xi\phi)+e_{15}\left[\frac{\partial}{\partial r}r\left(\frac{\partial U_{r}}{\partial z}\right)+\frac{\partial^{2}U_{\theta}}{\partial\theta\partial z}\right]/r=0;$$

$$\nabla^{2}(c_{55}EU_{z}+e_{15}\phi)+c_{55}E\left[\frac{\partial}{\partial r}r\left(\frac{\partial U_{r}}{\partial z}\right)+\frac{\partial^{2}U_{\theta}}{\partial\theta\partial z}\right]/r=0,$$

the general solution of which has the form

$$e_{15}U_z - e_{11}\varepsilon_{\varphi} = (Ar + B/r)\cos\theta; \quad c_{55}^E U_z + e_{15}\varphi = (Cr + D/r)\cos\theta; \\ U_r = Ez\cos\theta, \quad U_{\theta} = -Ez\sin\theta.$$
(18)

Here, $\nabla^2 = \frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial}{\partial r} \right) + \frac{1}{r^2} \frac{\partial^2}{\partial \theta^2}$ is the two-dimensional Laplace operator; $e_{15} = d_{15}c_{55}^{\ F}$; $c_{55}^{\ E} = (s_{55}^{\ E})^{-1}$; $\varepsilon_{11}^{\ \xi} = \varepsilon_{11}^{\ \sigma} (1 - k_{15}^{\ 2})$; $k_{15} = d_{15}\sqrt{c_{55}^{\ E}/\varepsilon_{11}^{\ \sigma}}$ electromechanical coupling factor. Having inserted (18) into (4), we find that the above solution satisfies the conditions of strain compatibility and electric-field potential. The unknown constants A, B, C, D, and E in the internal and external cylinders are determined from the boundary conditions on the surfaces of the compound cylinders.

Analysis of solution (18) showed that the success of the method of finding the effective constants from system of equations of state (2) by means of definitions of the type (8) depends on the procedure used to assign the boundary conditions at r = b and, moreover, that this method cannot be used to find the exact value. This is due to the fact that, in contrast to the previous case, the field of variables of the solution in (2) turns out to be highly nonuniform. Thus, we find variational estimates for the effective properties of the PCM by applying different types of boundary conditions and calculating the corresponding energy stored by the compound cylinder.

The boundary conditions at r = a will be continuity of the displacements and stresses, the tangential component of the electric field, and the normal component of induction:

$$U'_{r} = U''_{r}, \ U'_{z} = U''_{z}; \ \sigma'_{zr} = \sigma''_{zr}; \ E'_{\theta} = E''_{\theta}; \ D'_{r} = D''_{r}.$$
(19)

The boundary conditions at r = b will determine the corresponding variational boundary for the effective constant.

Let us find the lower variational boundary for the permittivity of a mechanically free compound cylinder $\ell \varepsilon_{11}^{*\sigma}$. We assign the electric displacement D* in the plane of isotropy of the PCM. Then at r = b

$$\sigma''_{zr} = 0; \quad D''_{r} = D^* \cos \theta. \tag{20}$$

The electrostatic energy stored by the compound cylinder is equal to

$$U = \frac{1}{2} \int_{V} \mathbf{D} \mathbf{E} dV = -\frac{1}{2} \int_{S} D''_{r} \varphi'' dS = -\frac{b}{2} \int_{0}^{l} dz \int_{0}^{2\pi} D''_{r} \varphi'' d\theta.$$
(21)

Having calculated $\varphi''(D^*)$ by means of (18)-(20) and having inserted the result into (21), we find the lower variational limit for the effective permittivity by comparing U(D^{*}) (21) with the energy of an equivalent homogeneous cylinder with the effective properties: U(D^{*}) = $D^{*2}V_c/2b\epsilon_{11}^{*\sigma}$. Here, $V_c = \pi b^2 \ell$ is the volume of the cylinder.

To determine the upper variational boundary $u\epsilon_{11}^{*\sigma}$, it is necessary to assign the electric potential at r = b (the field $E^* = -\varphi^*/b$):

$$\sigma''_{zr} = 0; \quad \varphi'' = \varphi^* \cos \theta \quad (E''_{\theta} = E^* \sin \theta). \tag{22}$$

Then having used (18), (19), and (22) to determine $D''_{r}(\varphi^{*})$ and having inserted the result into (21), we find $u\epsilon_{11}^{*\sigma}$ by comparing $U(\varphi^{*})$ (21) with the energy of an equivalent homogeneous cylinder with effective properties: $U(\varphi^{*}) = u\epsilon_{11}^{*\sigma} E^{*2}V_{c}/2$. It turns out that the upper and lower estimates of $\epsilon_{11}^{*\sigma}$ coincide and thus determine the exact value of permittivity in the PCM model being examined:

$$\varepsilon_{11}^{*\sigma} = \varepsilon_{11}^{''\sigma} \frac{(\varepsilon_{11}^{'\sigma} + \langle \varepsilon_{11}^{\sigma} \rangle) (c_{55}^{'E} + \langle c_{55}^{E} \rangle) - m_2 (1 + m_1) c_{55}^{'E} c_{55}^{''E} \tilde{d}_{15}^2}{(\varepsilon_{11}^{''\sigma} + [\varepsilon_{11}^{\sigma}]) (c_{55}^{'E} + \langle c_{55}^{E} \rangle) - m_2^2 c_{55}^{'E} c_{55}^{''E} \tilde{d}_{15}^2}.$$
(23)

The elastic energy stored up by the compound cylinder with the imposition of mechanical boundary conditions in the state of simple shear $(U''_z(b) = 0)$ is determined in the form

$$W = \frac{1}{2} \int_{V} \sigma_{ij} \xi_{ij} dV = \frac{1}{2} \int_{S} \sigma_{ij} n_{j} U_{i} dS = \frac{b^{2}}{4} \int_{0}^{1} dz \int_{0}^{2\pi} (\sigma''_{rz} U''_{r} + \sigma''_{\theta z} U''_{\theta}) \big|_{r=0} d\theta.$$
(24)

We assign the following mechanical and electrical boundary conditions at r = b:

$$U''_{z}=0; \quad D''_{r}=0; \quad \sigma''_{zr}=\sigma^{*}\cos\theta.$$
 (25)

Calculating W(σ^*) by means of (18), (19), and (25) with allowance for the symmetry of the stress tensor and the equilibrium equations and comparing the result with the energy of an equivalent homogeneous cylinder with effective properties (W(σ^*) = σ^{*2} V_c/2·_bc₅₅*^D), we find the lower variational estimate of the effective elastic modulus $\ell_{c_{55}}$ *^D in the no-load regime.

By assigning boundary conditions for the displacements at r = b

$$U''_{z}=0; \quad D''_{r}=0; \quad U''_{r}=\xi^{*}z\cos\theta, \quad U''_{\theta}=-\xi^{*}z\sin\theta,$$
(26)

corresponding to simple shear $(|\xi''_{ZT}(b)| = |\xi''_{Z\theta}(b)| = \xi^*/2)$, we can find the upper variational boundary for the effective elastic modulus ${}_{uC_{55}} {}^{*D}$ by comparing the elastic energy of the compound cylinder W(ξ^*) (24) calculated from (18), (19), and (26) with the energy of an equivalent homogeneous cylinder with effective properties: W(ξ^*) = ${}_{uC_{55}} {}^{*D}\xi^{*2}V_{c}/2$. Calculations of W(σ^*) and W(ξ^*) leads to coincidence of the upper and lower variational estimates, which establishes the exact value of the effective shear modulus in the no-load regime

$$c_{55}^{*D} = c_{55}^{''D} \frac{(c_{55}^{'E} + \langle c_{55}^{E} \rangle) (\varepsilon_{11}^{'\sigma} + \langle \varepsilon_{11}^{\sigma} \rangle) - m_2 (1 + m_1) c_{55}^{'E} c_{55}^{''E} \vec{d}_{15}^2}{\{(c_{55}^{''E} + [c_{55}^{E}]) (\varepsilon_{11}^{'\sigma} + \langle \varepsilon_{11}^{\sigma} \rangle) - m_2^2 c_{55}^{'E} \vec{c}_{55}^{''E} \vec{d}_{15}^2 - -4m_1 \varepsilon_{11}^{'\sigma} c_{55}^{''D} (k_{15}^{'2} - k_{15}^{''2})\}}$$
(27)

Calculation of the elastic energy stored in the compound cylinder with mechanical boundary conditions (25) and (26) in the SC regime $(E''_{\theta}(b) = 0)$, and its subsequent estimation by the above procedure also yield coincident variational boundaries, i.e., the effective elastic modulus

$$c_{55}^{*E} = c_{55}^{''E} \frac{(c_{55}^{'E} + \langle c_{55}^{E} \rangle) (\varepsilon_{11}^{''\sigma} + [\varepsilon_{11}^{\sigma}]) - m_2^2 c_{55}^{'E} c_{55}^{''E} \tilde{d}_{15}^2}{(c_{55}^{''E} + [c_{55}^{E}]) (\varepsilon_{11}^{''\sigma} + [\varepsilon_{11}^{\sigma}]) - m_2 (1 + m_1) c_{55}^{'E} c_{55}^{''E} \tilde{d}_{15}^2}.$$
(28)

The electrical state of the medium does not affect the shear modulus in the plane of isotropy $c_{66}^{*} = (c_{11}^{*E} - c_{12}^{*E})/2$, and it can be calculated from the solution of the elastic problem. Variational estimates for it were given in [6], while an exact solution was presented in [5] within the framework of a three-phase model of a medium with cylindrical inclusions.

To determine the effective piezomodulus d_{15}^{*} under conditions of simple shear $(U'_{z}(b) = 0)$, we examine the following boundary conditions at r = b:



Fig. 1. Effective values of the piezomodulus d_{33}^* (a), bulk piezomodulus d_V^* (b), and piezoelectric sensitivity $g_V^* = d_V^* / \epsilon_{33} d_C^*$ (c) of a PCM in relation to the concentration m_1 of PZT-5 piezoceramic fibers for three types of fillers (with $\epsilon'' / \epsilon_0 = 5$): 1) an elastomer with a Young' modulus E = 0.3GPa and Poisson's ratio $\nu = 0.45$ [12]; 2) epoxy resin with E = 3 GPa and $\nu = 0.4$ [12]; 3) boron-silicate glass with E = 74 GPa and $\nu = 0.21$ [13].

$$E''_{\theta} = E^* \sin \theta; \quad \sigma''_{ij} = 0. \tag{29}$$

The total electric energy stored up by the compound cylinder is equal to $U = \varepsilon_{11}^{*\sigma} E^{*2} V_C/2$ (23). As a result of the piezoeffect, part of this energy is converted to mechanical energy equal to $W = k_{15}^{*2} \varepsilon_{11}^{*\sigma} E^{*2} V_C/2$. The upper variational boundary for this energy can be found through the maximum strain of the compound cylinder with effective properties (28). It is clear from (2) that this value is equal to $2\xi''_{\theta Z}(b)$, averaged over the angle θ , since - by virtue of the second condition of (29) - it corresponds to the condition of free expansion of the cylinder, i.e.,

$$\frac{1}{2} k_{15}^{*2} \epsilon_{11}^{*\sigma} E^{*2} V_c \leqslant \frac{1}{2} c_{55}^{*E} \langle 2\xi''_{\theta z}(b) \rangle^2 V_c.$$

We can use this expression to obtain the upper variational limit for the piezomodulus $(k_{15}^{*2} = d_{15}^{*2}c_{55}^{*E}/\epsilon_{11}^{*\sigma})$:

$$\langle 2\xi''_{\theta z}(b) \rangle = d^*_{15} E^*. \tag{30}$$

However, on the other hand, for an equivalent homogeneous cylinder with the same boundary conditions, $2\xi^*_{\theta Z} = d^*_{15}E^*$, i.e., $\langle 2\xi''_{\theta Z}(b) \rangle = 2\xi^*_{\theta Z}$. This means that Eq. (30) gives the exact value of the effective piezomodulus (since the strain $\langle 2\xi''_{\theta Z}(b) \rangle$ is connected with redistribution of the piezostresses in the compound cylinder between the components of the PCM by virtue of (19), then its result will be due to the effective properties of the compound cylinder). With allowance for (18), (19), and (29), Eq. (30) gives

$$d^{*}_{15} = d''_{15} + 4m_{1}c_{55}'^{E}\epsilon_{11}''^{\sigma}\tilde{d}_{15} / [(c_{55}'^{E} + \langle c_{55}^{E} \rangle)(\epsilon_{11}''^{\sigma} + [\epsilon_{11}^{\sigma}]) - m_{2}^{2}c_{55}'^{E}c_{55}''^{E}\tilde{d}_{15}^{2}].$$
(31)

Thus, for the boundary conditions $D''_r(b) = D^* \cos \theta$ and $\sigma''_{ij}(b) = 0$, we calculate the effective piezoelectric sensitivity $g^*_{15} = \langle 2\xi''_{rz}(b) \rangle / D^*$.

The results obtained for the effective piezoconstants agree with the calculations of the dielectric (23) and elastic properties (27), (28) of the PCM, which follows from the relations in [7] for homogeneous piezoelectric materials: $g_{15}^* = d_{15}^*/\epsilon_{11}^{*\sigma}$; $d_{15}^* = \sqrt{\epsilon_{11}^{*\sigma}(S_{55}^{*E} - S_{55}^{*D})}$.

It should be noted that a similar relation $(\xi^*_{\theta} = \xi^{"}_{\theta}(b))$ between the strain in a medium with effective properties ξ^*_{θ} and the strain of the external coaxial cylinder $\xi^{"}_{\theta}(b)$ follows from the continuity of the displacement component U_r at $r = b(U^*_r = U^{"}_r)$ in the self-consistent field method when the effective piezomodulus $d^*_{31} = \xi^*_{\theta}/E^*_z$ is calculated. Then, with allowance for (7), we can calculate d^*_{31} from the relation $d^*_{31} = \xi^{"}_{\theta}(b)/E^*_z$ without integrating. The result coincides with (11), which once again validates the proposed approximations involved in the selection of the theoretical PCM mode.

4. Figure 1 shows the dependence of the piezomodulus d_{33}^* , the bulk piezomodulus d_V^* , and the piezoelectric sensitivity g_V^* of the PCM on the concentration of PZT-5 piezoceramic rods [7] for several fillers. The anomalies seen in piezoelectric sensitivity g_{31}^* , g_{33}^* , g_V^* , and the increase which occurs in c_{55}^{*E} and the piezocoefficients h_{33}^* , h_V^* , e_{33}^* , e_V^* in the PCM compared to the homogeneous piezoceramic can be attributed to the appreciable redistribution of the mechanical and electric fields in the nonuniform piezotexture from one component to the next. This is quite evident from the expressions for the mean fields. For example, the anomaly of g_{33}^* (and, accordingly, g_V^*) is readily discerned by comparing the dependences of d_{33}^* and $\epsilon_{33}^{*\sigma}$ on m_1 . Thanks to the high degree of compliance of the matrix, the stress applied in the measurement of g_{33}^* is taken up by the more rigid PZT. The latter for the most part also detemines the effective piezomodulus ($\approx d_{33}^*$), despite the "dilution" of the ceramic at $m_1 < 1$. The effective permittivity of such a PCM decreases sharply, which also accounts for the anomaly of g_{33}^* with the retention (up to $m_1 \approx 20\%$) of a substantial value of $d_{33}^* \approx d_{33}^*$.

Thus, the set of matrices obtained for the effective properties of a transversely isotropic PCM - (9-11), (13), (14-17), (23), (27), (28), and (31) - makes it possible to analyze and predict the properties of piezocomposites. This in turn makes it easier to solve one of the most basic problems in materials engineering - develop piezoelectric materials with prescribed properties.

LITERATURE CITED

- M. J. Haun, P. Moses, T. R. Gururaja, W. A. Schulze, and R. E. Newnham, "Transversal reinforced 1-3 and 1-3-0 piezoelectric composites," Ferroelectrics, <u>49</u>, No. 1-4, 259-264 (1983).
- H. Banno, "Theoretical equations for dielectric and piezoelectric properties of ferroelectric composites based on modified cubes model," Jpn. J. Appl. Phys., <u>24</u>, Pt. 1, Suppl. 24-2, 445-447 (1984).
- 3. A. A. Grekov, S. O. Kramarov, and A. A. Kuprienko, "Anomalous behavior of the twophase lamellar piezoelectric texture," Ferroelectrics, <u>76</u>, No. 1-2, 43-48 (1987).
- L. P. Khoroshun and B. P. Maslov, "Principles of a theory for predicting the electromechanical properties of heterogeneous piezoelectric materials," Prikl. Mekh., <u>15</u>, No. 8, 3-7 (1979).
- 5. R. Christensen, Introduction to the Mechanics of Composites [Russian translation], Moscow (1982).
- Z. Hashin and B. W. Rosen, "The elastic moduli of fiber-reinforced materials," J. Appl. Mech., <u>31</u>, No. 2, 223-232 (1964).
- 7. D. Berlincur, D. Curran, and G. Jaffe, "Piezoelectric and piezomagnetic materials and their use in transducers," Fiz. Akustika, 1, Pt. A, 204-326 (1966).

- 8. L. N. Lifschitz and L. N. Rozentsveig, "Toward a theory of the elastic properties of polycrystals," Zh. Éksp. Teor. Fiz., 16, No. 11, 967-980 (1946).
- 9. L. D. Landau and E. M. Lifschitz, Continuum Electrodynamics, Moscow (1982).

10. Yu. I. Sirotin and M. P. Shaskol'skaya, Principles of Crystal Physics, Moscow (1979).

- 11. Yu. N. Rabotnov, Mechanics of Deformable Solids [in Russian], Moscow (1979).
- 12. Encyclopedia of Polymers, Vol. 2, pp. 230-241 (1974); Vol. 3, p. 315 (1977), Moscow.
- 13. N. M. Pavlushkina (ed.), Glass (Handbook) [in Russian], Moscow (1973).

THERMAL EXPANSION OF A POLYMER COMPOSITE WITH AN AGGREGATING DISPERSE FILLER

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UDC 539.376:678.067

There are few experimental [1-4] and theoretical [5-11] studies of thermal expansion of polymer composites with rigid disperse fillers. Ideal dispersion of the filter in the material and the absence of contacts between inclusions are assumed in most of them. However, it is known that aggregation of particles of the disperse filler can have a significant effect on the effective properties of the composites. Effects of an increase in the viscosity [12, 13], electric and thermal conductivity [14-16], thermostability [17], and dynamic stiffness [18, 19] of composites based on thermoelastic and thermoreactive binders caused by agglomeration of particles are described in the literature. The possibility of a decrease in the thermal expansion coefficients of filled materials due to aggregation is noted in [1, 20]. It was found in [21, 22] that the effect of aggregation results in a significant increase in the modulus of elasticity and creep inhibition in high-density polyethylene (HDPE) filled with calcite. The presence of aggregation of the mineral filler in a composite has been confirmed experimentally. The features of thermal expansion of HDPE with an aggregating filler were experimentally studied in a wide range of temperatures and the possibilities of predicting the effective thermal expansion coefficient of similar composite materials (CM) were also investigated in the present study.

1. HDPE filled with activated calcite, whose deformation properties were studied in [21, 22], were tested. The samples, in the form of $50 \times 10 \times 3$ mm parallelepipeds, were cut



Fig. 1. Temperature dependences of deformation of a composite based on HDPE with v = 0 (1); 0.08 (2); 0.16 (3); 0.26 (4); 0.34 (5).

Institute of Polymer Mechanics, Academy of Sciences of the Latvian SSR, Riga. Translated from Mekhanika Kompozitnykh Materialov, No. 1, pp. 70-77, January-February, 1989. Original article submitted Septermber 16, 1988.