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2013

Della, C. N., & Shu, D. (2013). Performance of 1–3 piezoelectric composites with porous piezoelectric matrix. *Applied physics letters*, 103(13), 132905-.

<https://hdl.handle.net/10356/85684>

<https://doi.org/10.1063/1.4822109>

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Citation: [Applied Physics Letters](#) **103**, 132905 (2013); doi: 10.1063/1.4822109

View online: <http://dx.doi.org/10.1063/1.4822109>

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Performance of 1–3 piezoelectric composites with porous piezoelectric matrix

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(Received 25 April 2013; accepted 8 September 2013; published online 24 September 2013)

A micromechanics method is developed to investigate the effects of porosity in the matrix and the polarization orientations of the piezopolymer matrix and the piezoceramic fibers on the performance of 1–3 piezoelectric composites. The Mori-Tanaka (MT) method is first used to homogenize the porous piezopolymer matrix, and then the MT method for piezoelectric composites is used to analyze the porous piezopolymer matrix with embedded piezoceramic fibers. Results show that the performance of the composites is significantly enhanced by the presence of pores in the matrix and further enhanced by the opposite polarization orientation of the matrix and the fibers. © 2013 AIP Publishing LLC. [<http://dx.doi.org/10.1063/1.4822109>]

Piezoelectric composites or piezocomposites with 1–3 connectivity are an important class of materials for biomedical imaging and underwater acoustic applications.¹ These piezocomposites are flexible to conform to curved surfaces and have lower acoustic impedance and higher electromechanical coupling in comparison with single phase piezoceramic materials.

Analytical models^{2–10} and finite element models^{11–14} have been presented to study 1–3 piezocomposites with a non-porous matrix. Simple models for composites used in ultrasonic transducer application have been presented by Smith and co-workers^{2–4} and Chan and Unsworth.⁵ Nan and Weng⁶ presented a model using the effective medium theory for composites with piezopolymer matrix and piezoceramic fibers that was used to investigate the influence of polarization orientation of the piezoelectric matrix and fibers on the performance of the composites. Kar-Gupta and Venkatesh⁷ presented an equivalent layer composite model for composites where both the matrix and the fibers are anisotropic and piezoelectric active. Dunn and Taya^{8,9} and Odegard¹⁰ developed analytical models using the self-consistent, Mori-Tanaka (MT), and differential micromechanics theories to predict the effective electromechanical properties of piezocomposites.

Further studies have shown that the additional porosity in the matrix,^{15–19} modification of the poling orientations of the piezoelectric matrix and fibers,^{6,12,20} and the use of a flexible piezoelectric matrix²⁰ can enhance the performance of the 1–3 piezocomposites. Thus, motivated by these findings, the objectives of the present study are: (1) to develop a micromechanics-based method to analyze the performance of 1–3 piezocomposites with a porous piezopolymer matrix; and (2) to conduct a parametric study to examine the effects of the porosity in the piezopolymer matrix and the polarization orientations of the piezopolymer matrix and the piezoceramic fibers on the performance of 1–3 piezocomposites.

The micromechanics-based method for 1–3 piezocomposites presented here is a two-step process. First, the porous piezopolymer matrix is homogenized using MT model. Then, the porous matrix, which is assumed to be transversely isotropic, is embedded with piezoceramic fibers and analyzed using MT approach.^{8,9,16} The method is then used to study 1–3 piezocomposites for underwater acoustic transducer (hydrophone) and biomedical imaging applications, which consists of an array of parallel piezoceramic fibers embedded in a piezopolymer matrix, as shown in Fig. 1.

For linear piezoelectric materials, the constitutive equations are^{16,22}

$$\Sigma_{iJ} = E_{iJMn} Z_{Mn}, \quad (1)$$

where Σ_{iJ} , E_{iJMn} , and Z_{Mn} are

$$\Sigma_{iJ} = \begin{cases} \sigma_{ij} & J = 1, 2, 3 \\ D_i & J = 4, \end{cases} \quad (2)$$

$$E_{iJMn} = \begin{cases} C_{ijmn} & J, M = 1, 2, 3 \\ e_{nij} & J = 1, 2, 3; M = 4 \\ e_{imn} & J = 4; M = 1, 2, 3 \\ -\kappa_{in} & J, M = 4, \end{cases} \quad (3)$$

$$Z_{Mn} = \begin{cases} \epsilon_{mn} & M = 1, 2, 3 \\ -E_n & M = 4, \end{cases} \quad (4)$$

where σ_{ij} , ϵ_{ij} , E_i , and D_i are the stress tensor, strain tensor, electric field vector, and electric displacement vector, respectively. C_{ijmn} , e_{nij} , and κ_{in} are the elastic stiffness tensor, piezoelectric tensor, and permittivity tensor, respectively.

Equation (1) can be expressed in matrix form as

$$\Sigma = \mathbf{E} \mathbf{Z}, \quad (5)$$

where

$$\Sigma^T = [\sigma_{11} \quad \sigma_{22} \quad \sigma_{33} \quad \sigma_{23} \quad \sigma_{13} \quad \sigma_{12} \quad D_1 \quad D_2 \quad D_3], \quad (6)$$

$$\mathbf{Z}^T = [\epsilon_{11} \quad \epsilon_{22} \quad \epsilon_{33} \quad \gamma_{23} \quad \gamma_{13} \quad \gamma_{12} \quad -E_1 \quad -E_2 \quad -E_3], \quad (7)$$

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FIG. 1. (a) Schematic of a 1–3 piezoelectric composite with a porous piezopolymer matrix; (b) the polarization orientation of the piezopolymer matrix is assumed to be perfectly parallel (up or 0°) or antiparallel (down or 180°) with respect to the piezoceramic fiber. The porosity in the piezopolymer matrix occurs in a much finer length scale than the size of the piezoceramic fibers.

$$\mathbf{E} = \begin{bmatrix} \mathbf{C} & \mathbf{e}^t \\ (6 \times 6) & (6 \times 3) \\ \mathbf{e} & -\boldsymbol{\kappa} \\ (3 \times 6) & (3 \times 3) \end{bmatrix}, \quad (8)$$

and where $\mathbf{C}$, $\mathbf{e}$, and $\boldsymbol{\kappa}$ denote the elastic stiffness matrix, the piezoelectric constant matrix, and the permittivity matrix, respectively.

The MT method has been used by many to model a wide range of composites.^{23,24} For a porous piezopolymer matrix, the effective elastic moduli using the MT method are given by

$$\mathbf{E}^m = \mathbf{E}^p + v_v \langle (\mathbf{E}^v - \mathbf{E}^p) \mathbf{A}^m \rangle [(1 - v_v) \mathbf{I} + v_v \langle \mathbf{A}^m \rangle]^{-1}, \quad (9)$$

where $\mathbf{E}^m$, $\mathbf{E}^p$, $\mathbf{E}^v$, v_v , and $\mathbf{A}^m$ denotes the effective electroelastic moduli of the porous piezopolymer matrix, elastic electroelastic moduli of the matrix, elastic electroelastic moduli of the pores, volume fraction of the pores, and dilute concentration tensor. For the pore, elastic modulus is zero, $\mathbf{E}^v = 0$. The dilute concentration tensor $\mathbf{A}^m$ is given by

$$\mathbf{A}^m = [\mathbf{I} + \mathbf{S}^e (\mathbf{E}^p)^{-1} (\mathbf{E}^v - \mathbf{E}^p)]^{-1}, \quad (10)$$

where $\mathbf{S}^e$ is the elastic Eshelby tensor.^{23,24}

To calculate the effective electromechanical properties of the 1–3 piezoelectric composites, we assume the following: (a) the porous piezopolymer matrix is transversely isotropic; (b) the permittivity $\boldsymbol{\kappa}$ of the porous matrix is equal to

that of the dense matrix. This approximation results in a small error since $\boldsymbol{\kappa}$ is relatively small;¹⁵ and (c) the size of the pores is much less than the size of the piezoceramic fibers and so the fibers can be considered to be perfectly bonded to the porous piezopolymer matrix.²⁵

Using the MT method, the effective electroelastic moduli of piezoelectric composites are

$$\bar{\mathbf{E}} = \mathbf{E}^m + v_f \langle (\mathbf{E}^f - \mathbf{E}^m) \mathbf{A}^f \rangle [(1 - v_f) \mathbf{I} + v_f \langle \mathbf{A}^f \rangle]^{-1}, \quad (11)$$

where $\bar{\mathbf{E}}$ is the effective electroelastic moduli of the composite, $\mathbf{E}^m$ is the electroelastic moduli of the porous matrix, $\mathbf{E}^f$ is the electroelastic moduli of the piezoelectric fiber, v_f is the volume fraction of the piezoelectric fiber, $\mathbf{A}^f$ is the dilute concentration tensor and where the mechanical properties of the reference material (porous piezopolymer matrix) are those determined using Eq. (1). The dilute mechanical concentration tensor $\mathbf{A}^f$ is

$$\mathbf{A}^f = [\mathbf{I} + \mathbf{S}^p (\mathbf{E}^m)^{-1} (\mathbf{E}^f - \mathbf{E}^m)]^{-1}, \quad (12)$$

where $\mathbf{S}^p$ is the piezoelectric Eshelby tensor.^{9,24}

The hydrostatic parameters that are used to measure the performance of underwater acoustic transducers or hydrophones are: (a) hydrostatic charge coefficient d_h ; (b) hydrostatic voltage coefficient g_h ; and (c) hydrophone figure of merit (FOM) $d_h g_h$. These parameters can be determined by using the calculated effective electroelastic properties using Eq. (11), and the higher the value of each of these parameters, the higher is the sensitivity of the transducer.¹⁷ The hydrostatic charge coefficient d_h is defined by $d_h = d_{31} + d_{32} + d_{33}$. Large values of d_h are desirable for hydrophones in order to achieve enhanced sensitivity to the detection of sound waves. The corresponding hydrostatic voltage coefficient g_h is defined by $g_h = d_h^2 / \bar{\kappa}_{33}$. A useful FOM is the product of d_h and g_h , which should be maximized in order to enhance the signal-to-noise ratio of the transducer.²¹

For biomedical imaging applications, the electromechanical coupling k_t and the acoustic impedance Z are used to measure the performance of medical ultrasonic imaging transducers, where a low acoustic impedance and high electromechanical coupling is desired.⁴ The electromechanical coupling k_t is defined as $k_t = \sqrt{1 - \bar{C}_{33}^D / \bar{C}_{33}^D}$, where $\bar{C}_{33}^D = \bar{C}_{33} + (\bar{e}_{33})^2 / \bar{\kappa}_{33}$, and the acoustic impedance Z is given by $Z = (\bar{C}_{33}^D \rho)^{1/2}$, where the $\rho = v_f \rho_f + (1 - v_f) \rho_m$ and where ρ_f is the density of the fiber and ρ_m is the density of the porous matrix.

The 1–3 piezocomposites with cylindrical pores oriented along the x_3 -axis consists of PZT7A fibers and P(VDF-TrFE) matrix,^{16,20} as shown in Fig. 1. The fibers are aligned and poled along the x_3 -direction. The results for 1–3 piezocomposites with a non-piezoelectric Araldite D matrix¹⁶ are also presented for purpose of comparison. When the non-piezoelectric matrix is dense (non-porous) the results are identical to the results using the micromechanics model by Dunn and Taya.⁹

Figures 2(a)–2(c) show the effective hydrostatic parameters d_h , g_h , and $d_h g_h$ with respect to the piezoceramic fiber

FIG. 2. Effective hydrostatic performance parameters of 1–3 piezoelectric composites with a porous matrix as a function of the piezoceramic fiber (PZT-7A) volume fraction v_f for 0° and 180° polarization orientations of the piezopolymer matrix [P(VDF–TrFE)]: (a) Effective hydrostatic charge coefficient d_h ; (b) effective hydrostatic voltage coefficient g_h ; (c) effective hydrostatic figure of merit $d_h g_h$. When the Araldite D matrix is dense, i.e., 0% porosity, the results are identical to the results generated using the micromechanics model by Dunn and Taya.⁹

FIG. 3. (a) Effective electromechanical coupling k_t of 1–3 piezoelectric composites with a porous matrix as a function of the piezoceramic fiber (PZT-7A) volume fraction v_f for 0° and 180° polarization orientations of the piezopolymer matrix [P(VDF–TrFE)]; (b) effective electromechanical coupling k_t vs the acoustic impedance Z . When the Araldite D matrix is dense, i.e., 0% porosity, the results are identical to the results generated using the micromechanics model by Dunn and Taya.⁹

volume fraction v_f of the 1–3 PZT/P(VDF–TrFE) piezocomposites with cylindrical pores. The results for piezocomposites with parallel (0°) and antiparallel (180°) orientations of the piezopolymer matrix with respect to the piezoceramic fiber and with 0%, 10%, 20%, and 30% volume of porosity in the matrix are presented. It can be seen that the peak values of d_h , g_h , and $d_h g_h$ are much greater for the piezocomposites with piezoelectric polymer P(VDF–TrFE) matrix than for the piezocomposites with non-piezoelectric polymer Araldite D matrix.¹⁶ Furthermore, these parameters increase as the volume fraction of the porosity increase for up to $v_f < 0.3$. The polarization orientation has a more pronounced effect on the effective parameters of the piezocomposites for up to $v_f < 0.3$. The peak values of the effective parameters d_h , g_h , and $d_h g_h$ being higher for the antiparallel orientation (180°) and attained at lower volume fraction of the piezoceramic fibers as compared with the results for the parallel orientation (0°).

The electromechanical coupling k_t of the composites with respect to the fiber volume fraction v_f is shown in Fig. 3(a). It is observed that k_t increases as the volume fraction of the porosity increases, with k_t being higher for the antiparallel orientation. The effective electromechanical coupling k_t vs the effective acoustic impedance Z for the piezoelectric composites is shown in Fig. 3(b). It can be seen that k_t first increases rapidly with Z and approaches an asymptotic value after $Z = 6$ and that k_t further increases as the volume of porosity increases. The composites with antiparallel orientation reach higher k_t values at lower Z , as compared with the composites with parallel orientation.

In summary, the presence of porosity enhanced the effective hydrostatic performance parameters of the 1–3 piezocomposites by increasing the peak values of these parameters, since the peak values increased as the volume of porosity increased. The peak values were observed to be higher and attained at lower fiber volume ratio for composites with piezopolymer matrix and piezoceramic fibers poled in the opposite directions than composites with matrix and fibers poled in the same direction. The porosity in the piezopolymer matrix also improved the performance of the 1–3 piezocomposites by increasing the electromechanical coupling while reducing the acoustic impedance of the composite. The electromechanical coupling increased as the volume of the porosity in the matrix is increased and higher

electromechanical coupling of the composites are attained when the piezopolymer matrix and piezoceramic fibers are poled in the opposite directions.

We thank the anonymous reviewer for the valuable contributions.

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- ²⁵Note that Eq. (9) enables us to evaluate the electroelastic moduli of the porous piezoelectric matrix, and then these constants are used at averaging to obtain the effective electromechanical properties of the piezoelectric composite as a whole. In this case, the size (diameter) of the pores is much less than the piezoceramic fibers. These fibers are embedded into the homogeneous medium and do not “feel” a single pore.