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Effective properties of periodic or non-periodic multilayered thermopiezoelectric composites

Esperanza García Santoyo[a], Leonardo Cortés Carrasquero[a] , Raúl Martínez Rosado[b], Federico J. Sabina[c], Reinaldo Rodríguez Ramos[a], Julián Bravo Castillero[a].

[a] Facultad de Matemática y Computación, Universidad de La Habana. San Lázaro y L, Vedado, Habana 4, CP 10400, Cuba, jbravo@matcom.uh.cu
[b] Instituto Superior de Estudios Superiores de Monterrey, Campus Estado de México, Escuela de Ingeniería y Computación, Apartado Postal 52-926, Estado de México, México.
[c] Instituto de Investigaciones en Matemáticas Aplicadas y Sistemas, Universidad Nacional Autónoma de México. Delegación Álvaro Obregón, México 01000, D. F., México.

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Sumario. Aplicando el Método de Homogeneización Asintótica se obtienen fórmulas analíticas exactas para todas las propiedades efectivas de compuestos termo-piezoeléctricos formados por cualquier número finito de láminas. Se muestran cálculos numéricos para ilustrar la relevancia de estos resultados en aplicaciones al diseño de transductores para imágenes biomédicas e hidrófonos.

Abstract. Exact analytical formulae for the effective properties of periodic multilayered thermopiezoelectric composites are derived using the asymptotic homogenization method. The general expressions are also valid for plate laminated composites, i.e., a plate bounded by traction-free plane surfaces. Numerical calculations are shown to illustrate the relevance of these results in transducer biomedical imaging and hydrophone applications.

Palabras clave. Piezoelectric composite materials, 77.84 Lf, Acoustic properties of solids, 62.65.+k.

1 Introduction

It is well known that study of stratified media leads to exact formulae for their effective coefficients. See, for instance, in recent references [1] and [3]. In ref. [4] the asymptotic homogenization method is applied to find general formulae of two-layered thermopiezoelectric composites. The asymptotic homogenization model is also applied in ref. [2] to obtain general formulae for overall properties of n-layered piezoelectric composites by a previous generalization of the results published in Chapter 5 of ref [8] for the purely elastic case. In Chapter 9 of ref [6] an extensive revision of several results in the prediction of effective properties of layered structures elastic, thermoelastic, thermoelectric and piezoelectric is presented, and other important references in this area are quoted there. In ref. [9] the problem of homogenization of a thermopiezoelectric composite is also mathematically treated by the method of two-scale asymptotic expansions. The theoretical contribution of this paper is the generalization of the formulae in ref [4] to the case of multilayered thermopiezolectric composites. Following the model of ref. [5] it is also proved that the generalized formulae are also valid for the not necessarily regular structures. From a practical point of a view the relevance of this effort is shown by means of examples of three-layered composites useful either in biomedical imaging or hydrophone applications. The dependence of the overall properties of a composite as function of the individual physical properties and of the specific application of interest is also illustrated in these numerical experiments. In several cases the improvement of global characteristics is remarkable for the case of three-layered composites in respect to their binary counterparts.

In section 2, the statement of the problem is given for a periodic multilayered thermopiezoelectric structure, and the local problems, which are required to be solved, are formulated using a compact notation. A simple derivation of the corresponding effective formulae is described. Such formulae are exactly those reported in ref. [4] for the two-layered thermopiezolectric composite. In section 3 deals with useful numerical calculations to optimize ultrasonic transducer applications. In section 4 some concluding remarks are summarized.

2 Effective coefficients and local problems

Let $\Omega \subset \Re^3$ be a bounded laminated thermopiezolectric composite made of cells which are periodically distributed along Ox_3 axis. Each cell may be made of any finite number of thermopiezolectric layers. The axis of material symmetry of each layer are parallel to each other and the x_3-axis is perpendicular to the layering. The tensors of elastic, thermoelastic, piezoelectric, dielectric and pyroelectric modules are denoted by $c_{ijkl}, \gamma_{ij}, g_{ijk}, \in_{ij}$ and λ_i respectively. Throughout this paper Latin indices take values 1, 2 and 3; and Greek indices run from 1 to 4. The summation convention is understood, but it is taken only over repeated lower case indices. Next, k_{ij} stands for the heat conductivity and $\beta = \frac{C_e}{T_0}$; C_e is the specific heat at constant strain per unit volume and T_0 is the reference (absolute) temperature. Let the material functions be Y-periodic functions, where $Y = \{y_3 : 0 \le y_3 \le 1\}$ is the unit periodic cell. Here $y_3 = \frac{x_3}{\alpha}$ is the local (fast) coordinate and $\alpha = \frac{l}{L}$ is the geometrical small parameter, which represents the radio between the characteristic length l of the periodic cell Y, and the characteristic length L of the whole domain Ω.

By using a compact notation, introduced in ref. [4], the effective coefficients can be written as follows:

$$\overline{C}_{\alpha\beta\mu\nu} = \left\langle C_{\alpha\beta\mu\nu} + C_{\alpha\beta\gamma 3} \frac{d_{\mu\nu}\chi_\gamma}{dy_3} \right\rangle \quad (1)$$

$$\overline{K}_{ij} = \left\langle K_{ij} + K_{i3} \frac{d_j\Theta}{dy_3} \right\rangle \quad (2)$$

where $C_{ijmn} {}^o c_{ijmn}, C_{4jmn} {}^o g_{jmn}, C_{4j4n} {}^o - \hat{I}_{jn}, C_{ij44} {}^o -\gamma_{ij}, C_{4j44} {}^o \lambda_j$

and $C_{4444} \equiv \beta$. The angular brackets define the average per unit length of the relevant quantity over the unit cell, that is, $\langle F \rangle = \frac{1}{|Y|}\int_Y F(y_3)dy_3$, where $|Y|$ denotes the length of the unit periodic cell Y. For simplicity, the following notation will be used: $\langle F \rangle = \int_0^1 F(y_3)dy_3$. Note that the local auxiliary functions ${}_{\mu\nu}\chi_\gamma$ and ${}_j\Theta$ pre-indexes $\mu\nu$ and j use to relate them to certain differential equations L below are solutions of the following local problems:

Problem ${}_{\mu\nu}L$: find ${}_{\mu\nu}\chi_\gamma$ being Y-periodic with $\langle {}_{\mu\nu}\chi_\gamma \rangle = 0$ such that

$$\frac{d}{dy_3}\left(C_{\alpha 3\gamma 3} \frac{d_{\mu\nu}\chi_\gamma}{dy_3} + C_{\alpha 3\mu\nu} \right) = 0 \,. \quad (3)$$

Problem ${}_jL$: find ${}_j\Theta$ being Y-periodic with $\langle {}_j\Theta \rangle = 0$ such that

$$\frac{d}{dy_3}\left(K_{3j} + K_{33} \frac{d_j\Theta}{dy_3} \right) = 0 \,. \quad (4)$$

The above problems are solved using the procedure appear in Chapter 5 of ref. [8], for the purely elastic case; obtaining the following expressions for derivatives of the local functions:

$$\frac{d_{\mu\nu}\chi_\gamma}{dy_3} = C^{-1}_{\gamma 3\delta 3}\left(\left\langle C^{-1}_{\delta 3\rho 3} \right\rangle^{-1} \left\langle C^{-1}_{\rho 3\theta 3} C_{\theta 3\mu\nu} \right\rangle - C_{\delta 3\mu\nu} \right)$$

$$\frac{d_j\Theta}{dy_3} = K^{-1}_{33}\left(\left\langle K^{-1}_{33} \right\rangle^{-1} \left\langle K^{-1}_{33} K_{3j} \right\rangle - K_{3j} \right) \quad (5,6)$$

In (5) and below, the exponent -1 denotes the indicated component of the 4x4 inverse matrix. Finally, substituting (5), (6) into (1), (2) respectively, the effective coefficients take the simple form

$$\begin{aligned} \overline{C}_{\alpha\beta\mu\nu} &= \left\langle C_{\alpha\beta\mu\nu} \right\rangle \\ &+ \left\langle C_{\alpha\beta\gamma 3} C^{-1}_{\gamma 3\delta 3} \right\rangle \left\langle C^{-1}_{\delta 3\rho 3} \right\rangle^{-1} \left\langle C^{-1}_{\rho 3\theta 3} C_{\theta 3\mu\nu} \right\rangle , \\ &- \left\langle C_{\alpha\beta\gamma 3} C^{-1}_{\gamma 3\delta 3} C_{\delta 3\mu\nu} \right\rangle \end{aligned} \quad (7)$$

$$\begin{aligned} \overline{K}_{ij} &= \left\langle K_{ij} \right\rangle + \left\langle K_{i3} K^{-1}_{33} \right\rangle \left\langle K^{-1}_{33} \right\rangle^{-1} \left\langle K^{-1}_{33} K_{3j} \right\rangle \\ &- \left\langle K_{i3} K^{-1}_{33} K_{3j} \right\rangle \end{aligned} \,. \quad (8)$$

It is interesting to point out that these formulae are also valid for a finite laminate not necessarily periodic, that is, a plate. The expressions of the local problems and effective coefficients are the same in both cases, with the particularity that for the non-periodic problem, the local functions take the value zero on opposed sides of the unit cell Y [5]. These boundary conditions are equivalent to the periodicity conditions for the present one-dimensional case.

After some algebraic manipulations it is verified that the effective constants in (7) and (8), for the particular case of a binary medium, coincide exactly with formulae (17) and (18) reported in ref. [4].

In the engineering literature this kind of layer distribution ($y \equiv y_3$) is known as "connectivity in series" (see for instance Figure 1) and the case corresponding to $y \equiv y_1$ (or y_2) is called "connectivity in parallel", see for instance ref. [7]. From (7) and (8), interchanging in these expressions the indices 3 and 2 (or 1), the corresponding formulae for parallel connectivity case can be obtained.

3 Improvement of physical characteristics

In order to show the importance of these results, we will consider the case of parallel connectivity where each periodic cell consists of three different homogenous phases: a polymer phase (medium 1) which is piezoelectrically inactive or active, the medium 2 is a TLZ5 piezoelectric ceramic, and the medium 3 is a VDF/TrFE piezoelectrically active polymer. Two applications of these composite materials will be shown: one of them is relevant to passive detectors subjected to hydrostatic conditions (such as hydrophones) and the other one is related to transducers for biomedical imaging applications. The basic effective coefficients for computing the main physical parameters for both applications can be derived from (7) (replacing 3 by 2, and assuming that each phase belongs to the class 6mm). They are finally given by the following formulae:

$$\bar{c}_{11} = \sum \lambda_i c_{11}^{(i)} + \left(\sum \lambda_i c_{12}^{(i)} / c_{11}^{(i)}\right)^2 \Big/ \left(\sum \lambda_i / c_{11}^{(i)}\right), - \left(\sum \lambda_i \left(c_{12}^{(i)}\right)^2 \Big/ c_{11}^{(i)}\right)$$

$$\bar{c}_{12} = \left(\sum \lambda_i c_{12}^{(i)} / c_{11}^{(i)}\right) \Big/ \left(\sum \lambda_i / c_{11}^{(i)}\right),$$

$$\bar{c}_{13} = \sum \lambda_i c_{13}^{(i)} + \left(\sum \lambda_i c_{12}^{(i)} / c_{11}^{(i)}\right)\left(\sum \lambda_i c_{13}^{(i)} / c_{11}^{(i)}\right) \Big/ \left(\sum \lambda_i / c_{11}^{(i)}\right), - \sum \lambda_i c_{12}^{(i)} c_{13}^{(i)} / c_{11}^{(i)}$$

$$\bar{c}_{22} = 1 \Big/ \left(\sum \lambda_i / c_{11}^{(i)}\right),$$

$$\bar{c}_{23} = \left(\sum \lambda_i c_{13}^{(i)} / c_{11}^{(i)}\right) \Big/ \left(\sum \lambda_i / c_{11}^{(i)}\right),$$

$$\bar{c}_{33} = \sum \lambda_i c_{33}^{(i)} + \left(\sum \lambda_i c_{13}^{(i)} / c_{11}^{(i)}\right)^2 \Big/ \left(\sum \lambda_i / c_{11}^{(i)}\right), - \left(\sum \lambda_i \left(c_{13}^{(i)}\right)^2 \Big/ c_{11}^{(i)}\right)$$

$$\bar{g}_{31} = \sum \lambda_i g_{31}^{(i)} + \left(\sum \lambda_i g_{31}^{(i)} / C_{11}^{(i)}\right)\left(\sum \lambda_i C_{12}^{(i)} / C_{11}^{(i)}\right) \Big/ \left(\sum \lambda_i / C_{11}^{(i)}\right), - \sum \lambda_i g_{31}^{(i)} C_{12}^{(i)} / C_{11}^{(i)}$$

$$\bar{g}_{32} = \left(\sum \lambda_i g_{32}^{(i)} / C_{11}^{(i)}\right) \Big/ \left(\sum \lambda_i / C_{11}^{(i)}\right),$$

$$\bar{g}_{33} = \sum \lambda_i g_{33}^{(i)} + \left(\sum \lambda_i g_{31}^{(i)} / C_{11}^{(i)}\right)\left(\sum \lambda_i C_{13}^{(i)} / C_{11}^{(i)}\right) \Big/ \left(\sum \lambda_i / C_{11}^{(i)}\right), - \sum \lambda_i g_{31}^{(i)} C_{13}^{(i)} / C_{11}^{(i)}$$

$$\bar{\hat{I}}_{33} = \sum \lambda_i \hat{I}_{33}^{(i)} + \sum \lambda_i \left(g_{31}^{(i)}\right)^2 \Big/ C_{11}^{(i)}, - \left(\sum \lambda_i g_{31}^{(i)} / C_{11}^{(i)}\right)^2 \Big/ \left(\sum \lambda_i / C_{11}^{(i)}\right)$$

$$\bar{\rho} = \sum \lambda_i \rho^{(i)} \qquad (9)$$

where $\sum$ denotes the summation for i from 1 to 3, $\lambda_i = |Y_i| / |Y|$ is the volumetric fraction of phase i (which represents the ratio between the length $|Y_i|$, of the region occupied by phase i, and the length $|Y|$ of the periodic unit cell Y), and ρ is the mass density.

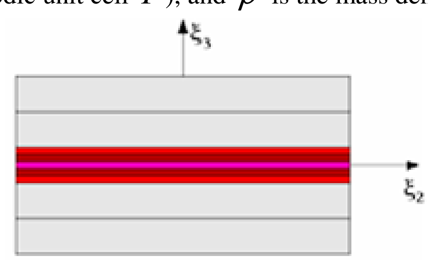

Figure 1: Layer distribution "connectivity in series"

The usual notation of adjacent indices: $((11) \to 1, (22) \to 2, (33) \to 3)$ has been used to express briefly the elastic, piezoelectric and dielectric coefficients.

Using the effective coefficients (9) it is possible to calculate the components of averaged tensors of elastic compliances $\bar{S}_{ij}^{E}$, piezomoduli $\bar{d}_{mi}$, elastic rigidity

$\bar{c}^{D}_{mn}$, and permittivity $\bar{\in}^{T}_{mn}$, by using the following formulae

$$\bar{S}^{E}_{ij} = (-1)^{i+j}\frac{\Delta_{ij}}{\Delta}, \qquad \bar{d}_{mi} = \bar{g}_{mj}\bar{S}^{E}_{ji},$$

$$\bar{c}^{D}_{mn} = \bar{c}_{mn} + \bar{g}^{2}_{mp}\bar{\in}^{-1}_{pn}, \qquad \bar{\in}^{T}_{mn} = \bar{\in}_{mn} + \bar{d}_{mp}\bar{g}_{pn},$$

where, Δ is the determinant of the $\bar{c}_{ij}$ matrix and Δ_{ij} is the minor obtained by excluding the ith row and jth column.

The use composite materials for hydrophone applications is based on the idea of decoupling the piezoelectric $\bar{d}_{33}, \bar{d}_{31}$ and $\bar{d}_{32}$ coefficients and lowering the permittivity $\bar{\in}^{T}_{33}$, these composites have produce some remarkable improvements in the hydrostatic $\bar{d}_{h}(= d_{31} + d_{32} + d_{33})$ and $\bar{g}_{h}\left(= \bar{d}_{h}\big/\left(\in_{0}\bar{\in}^{T}_{33}\right)\right)$ coefficients, where $\in_{0}$ denotes the permittivity of free space. The principle of designing a composite material for hydrophone applications is maintain $\bar{d}_{33}$ as large as possible and reduce $\bar{d}_{31}, \bar{d}_{32}$ and the dielectric constant $\bar{\in}^{T}_{33}$ resulting in an enhanced value of $\bar{d}_{h}\bar{g}_{h}$.

Figure 2 shows the variation of $\bar{d}_{h}\bar{g}_{h}$, the figure of merit, versus volume fraction of piezoelectric ceramic (λ_2) for four different three-layered composites, in parallel connection, made of a polymer (medium 1), TLZ5 (ceramic medium, 2) and VDF/TrFE (piezoelectric polymer, medium 3). The medium 1 is the only one varies for each composite. For composite 1, Eccothane (solid line) is used; and for composite 4, the medium 1 is the VDF/TrFE piezoelectric polymer (doted line) (in this case it becomes a two-layered composite). The material parameters used in the calculation are listed in Table 1. For the calculations the value of $\lambda_1 = 0.8$ is fixed. As it can be observed in Figure 1, the optimum percentage of TLZ5 to maximize the value of $\bar{d}_{h}\bar{g}_{h}$ should be about eight percent of TLZ5 for the best combination which is composite 2. Figure 3 illustrates the variation of thickness electromechanical coupling factor

$$\bar{K}_{t}\left(= \sqrt{1 - \bar{c}_{33}\big/\bar{c}^{D}_{33}}\right)$$

versus acoustic impedance $\bar{Z}\left(= \sqrt{\rho c^{D}_{33}}\right)$ for the same four composites involved in Figure 1. The objective now is to show which is the best combination in the design of transducer for biomedical imaging applications. In this case the piezoelectric material requires of a high electromechanical coupling coefficient (= 0.6 to 0.7) for high sensitivity; and a low acoustic impedance (Z < 7.5 Mrayl) to minimize reflection losses at the interfaces. The optimum material can be obtained by adjusting the volume fraction of ceramic piezoelectric. As the volume fraction decreases the acoustic impedance also diminished which eventually causes deterioration in the electromechanical coupling.

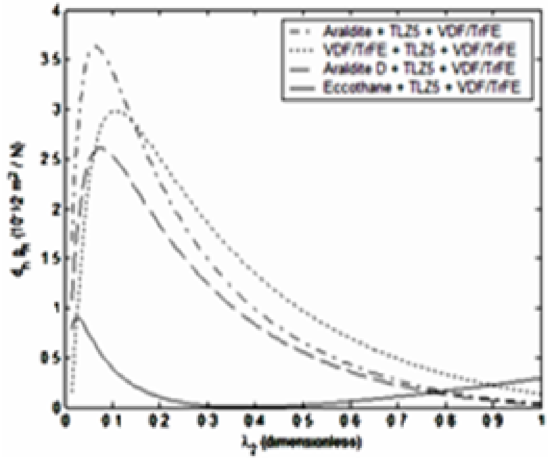

Figure 2. Figure of merit for hydrophone applications in three-layered piezocomposites, in parallel connection, between the products $\bar{d}_{h}\bar{g}_{h}$ versus volume fraction of piezoelectric ceramic TLZ-5.

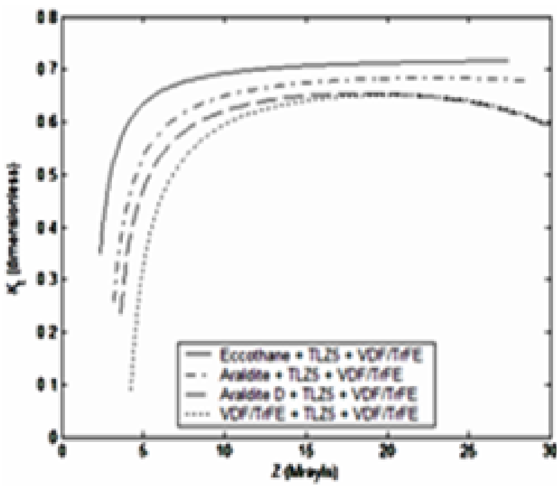

Figure 3. Trade-off between high electromechanical coupling and low acoustic impedance in three layered piezocomposites, in parallel connection, made from TLZ-5, VDF/TrFE and four different polymers.

A trade-off then must be made between minimizing the impedance and maximizing the coupling, as illustrated in this Figure the composite 1 reflects the best combination of materials for this application.

4 Concluding remarks

Analytical expressions of the effective coefficients for thermopiezoelectric layered composites with any finite number of layers are derived based on the asymptotic homogenization method (equations (7) and (8)). These general formulae are applied in order to calculate the overall thermopiezoelectric properties for layered media composed of layers with 6mm symmetry.

Table I
Material parameters used in the numerical examples; $\in_0 = 8.854.10^{-12}\, C^2/Nm^2$ is the free space permittivity.

Parameters	TLZ-5	VDF/TrFE	Araldite	Araldite D	Eccothane
$c_{11}\,(GPa)$	126	8.5	5.46	8.0	1.64
$c_{12}\,(GPa)$	79.5	3.6	2.94	4.4	1.57
$c_{13}\,(GPa)$	84.1	3.6	2.94	4.4	1.57
$c_{33}\,(GPa)$	109	9.9	5.46	8.0	1.64
$g_{31}\left(C/m^2\right)$	-6.5	0.008	0	0	0
$g_{33}\left(C/m^2\right)$	24.8	-0.29	0	0	0
$\in_{33}/\in_0$	1813	6.0	7.0	4.0	5.4
$\rho\left(kg/m^3\right)$	7898	1880	1170	1150	1130

Two examples of applications for the design of improved ultrasonic devices (hydrophones and biomedical imaging) are presented. The basic effective formulae for these applications (equations (9)) are of very easy computation. These examples illustrates that three layered composites can possess improved properties than two layered ones. The dependence of the overall properties of a composite relative to the physical characteristics of the individual phases and the predetermined application is also shown in these examples. For instance the combination Eccothane-TLZ5-VDF/TrFE is a very good candidate for biomedical imaging but is not recommendable for hydrophone applications.

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