

An Analytical Study on Prediction of Effective Properties in Porous and Non-Porous Piezoelectric Composites

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Eshelby type micromechanics model with a newly developed piezoelectric Eshelby tensor is proposed for predicting the effective electroelastic properties of the piezoelectric composite. The model is applied for piezoelectric solids containing both porosities and metal inhomogeneities. The effective electroelastic moduli of the composites such as stiffness, piezoelectric constants, and dielectric constants are predicted by the present model, which are extensively compared with the existing experimental results from the literatures. The validity of Eshelby type model for predicting the effective properties of the composite is thoroughly examined. It can be concluded from this study that a new mechanism is needed to compute correctly the dielectric constants among the effective properties of the composites.

Key Words : Piezoelectric Composite, Porous Composite, Non-Porous Composite, Effective Electroelastic Moduli, Piezoelectric Eshelby Tensor

Nomenclature

Σ : Stress and electric displacement
 L : Electroelastic moduli
 Z : Strain and electric field
 σ : Stress
 D : Electric displacement
 C : Elastic moduli
 e : Piezoelectric constants
 κ : Dielectric constants
 ϵ : Strain
 E : Electric field
 D : Piezoelectric composite domain
 \mathcal{Q} : Porosity or inhomogeneity domain
 S : Piezoelectric Eshelby tensor
 Σ^o : Applied uniform stress and electric displacement
 Z^o : Strain and electric field induced in the matrix without inhomogeneities by Σ^o

\bar{Z} : Average disturbance of the strain and electric field in the matrix
 Z : Disturbed strain and electric field in the inhomogeneities
 Z^* : Equivalent eigenstrain and electric field of equivalent inclusion
 Z_c : Total strain and electric field of the composite
 Z_m : Total strain and electric fields in the matrix
 Z_f : Total strain and electric fields in the inhomogeneities
 L_c : Effective electroelastic moduli of the composite
 v_f : Volume fraction of inhomogeneities

Subscripts

m : Matrix
 f : Inhomogeneities, porosity or filler
 c : Composite

1. Introduction

Piezoelectric composites have been widely used in the design of sensors and actuators such as sonar projector, underwater and medical imaging

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applications. Piezoelectric bimorph among piezoelectric actuators is well known to produce high bending displacement with high stress concentration at the interface between the top and bottom layers, reducing its longevity. In order to reduce the high stress concentration at the interface, the concept of functionally graded microstructure is introduced, where piezoelectric bimorph is composed of a number of laminae with a piezoelectric composite. In processing of piezoelectric materials, defects, cracks, or voids can occur naturally within materials. Porous piezoelectric ceramics such as lead zirconate titanate (PZT) offer significant improvements over solid piezoelectric ceramics in many piezoelectric transducer design figures of merit. The PZT/Pt composite shows the better fracture toughness than PZT only. By using these advantages for the application of piezoelectric ceramic as actuators, porosity-graded and PZT/Pt functionally graded piezoelectric actuators were fabricated (Li et al., 2001 ; 2003 ; Takaki et al., 2002 ; 2003 ; Taya et al., 2003). For the analysis of dynamic response of an anti-plane shear crack in functionally graded piezoelectric material (FGPM), the electroelastic material properties of the FGPM were assumed to vary smoothly in the form of an exponential function along the thickness of the strip (Kwon and Lee, 2004). Thus, a model predicting the effective electroelastic properties of piezoelectric solids containing both porosities and metal fillers is prerequisite to design piezoelectric composites.

Piezoelectric Eshelby tensors corresponding to Eshelby's tensor in elasticity (Eshelby, 1957) was derived to predict the effective elastic, dielectric, and piezoelectric moduli of piezoelectric composites (Dunn and Taya, 1993a ; Dunn and Wienecke, 1997 ; Huang and Yu, 1994 ; Michelitsch and Levin, 2000 ; Mikata, 2001). Mori-Tanaka theory (Mori and Tanaka, 1973) was extended to consider the coupled electroelastic behavior of piezoelectric ceramic reinforced polymer composites (Dunn and Taya, 1993b). By using piezoelectric Eshelby tensors and Mori-Tanaka theory, many researches for predicting the effective electroelastic properties of the piezoelectric composites have been undertaken for either porous (Dunn

and Taya, 1993c ; Wu, 2000) or non-porous composites (Dunn and Taya, 1993a ; 1993b ; Huang and Kuo, 1996 ; Kuo and Huang, 1997).

The electromechanical properties of porous piezoelectric ceramics have been predicted by coupling the exact solution for a single ellipsoidal pore embedded in an infinite piezoelectric matrix with an effective medium approximation, and compared with experimental results (Dunn and Taya, 1993c). The analytical models for predicting the effective electroelastic properties of piezoelectric matrix containing piezoelectric short fibers have been proposed, and showed numerical predictions for BaTiO₃/PZT-5H composite (Huang and Kuo, 1996 ; Kuo and Huang, 1997). Their predictions have not been justified by comparing experimental results. A unified micromechanics approach has been adopted to examine the effective electroelastic properties of porous piezoelectric composite (Wu, 2000). Effects of the volume fraction and aspect ratio of voids on the material properties have been studied, but no comparison was made with experimental results.

Although a number of models using Eshelby type approach have been proposed, no model has been applied for both the porous and non-porous piezoelectric composites at the same time and their limitations have not been demonstrated. In this study, the effective electroelastic properties of both the porous and non-porous piezoelectric composites are predicted by using Eshelby's equivalent inclusion method (Eshelby, 1957) with Mori-Tanaka's mean field theory (Mori and Tanaka, 1973). Among several piezoelectric Eshelby tensors (Dunn and Taya, 1993a ; Dunn and Wienecke, 1997 ; Huang and Yu, 1994 ; Michelitsch and Levin, 2000 ; Mikata, 2001), more explicit and newly derived piezoelectric Eshelby tensors are employed in this study (Mikata, 2001). Comparing the predicted results by the present model with existing experimental results for both porous piezoelectric ceramic (Li et al., 2003) and PZT/Pt piezoelectric composites (Li et al., 2001 ; Takaki et al., 2002 ; 2003 ; Taya et al., 2003), the validity of Eshelby type model for predicting the effective electroelastic moduli of piezoelectric composite is extensively discussed.

2. Formulation

The constitutive equations for linear piezoelectric materials are written in matrix notation as

$$\Sigma = LZ \tag{1}$$

where Σ , L , and Z represent the stress and electric displacement, electroelastic moduli, and the elastic strain and electric field matrices, respectively. Those matrices are composed of sub-matrices as

$$\Sigma = \begin{bmatrix} \sigma \\ D \end{bmatrix}, L = \begin{bmatrix} C & e \\ e & -\kappa \end{bmatrix}, Z = \begin{bmatrix} \varepsilon \\ -E \end{bmatrix} \tag{2}$$

where σ , D , C , e , κ , ε , and E denote stress, electric displacement, elastic moduli, piezoelectric constants, dielectric constants, strain, and electric field, respectively. Each sub-matrix for a transversely isotropic piezoelectric solid is given as follows

$$\begin{aligned} \sigma &= [\sigma_{11} \ \sigma_{22} \ \sigma_{33} \ \sigma_{23} \ \sigma_{13} \ \sigma_{12}]^T \\ \varepsilon &= [\varepsilon_{11} \ \varepsilon_{22} \ \varepsilon_{33} \ \varepsilon_{23} \ \varepsilon_{13} \ \varepsilon_{12}]^T \\ D &= [D_1 \ D_2 \ D_3]^T, E = [E_1 \ E_2 \ E_3]^T \\ \kappa &= \begin{bmatrix} \kappa_{11} & 0 & 0 \\ 0 & \kappa_{11} & 0 \\ 0 & 0 & \kappa_{33} \end{bmatrix}, e = \begin{bmatrix} 0 & 0 & 0 & 0 & e_{15} & 0 \\ 0 & 0 & 0 & e_{15} & 0 & 0 \\ e_{31} & e_{31} & e_{33} & 0 & 0 & 0 \end{bmatrix} \\ C &= \begin{bmatrix} C_{11} & C_{12} & C_{13} & 0 & 0 & 0 \\ C_{21} & C_{22} & C_{23} & 0 & 0 & 0 \\ C_{31} & C_{32} & C_{33} & 0 & 0 & 0 \\ 0 & 0 & 0 & C_{44} & 0 & 0 \\ 0 & 0 & 0 & 0 & C_{44} & 0 \\ 0 & 0 & 0 & 0 & 0 & C_{66} \end{bmatrix} \end{aligned} \tag{3}$$

where $C_{66} = (C_{11} - C_{12})/2$.

The composite of interest consists of an infinite piezoelectric matrix ($D-\Omega$) containing a finite volume fraction, v_f , of ellipsoidal inhomogeneities, as shown in Fig. 1(a). The ellipsoidal inhomogeneities of the same shape are considered as metals or porosities and aligned with the x_3 axis. The non-piezoelectric inhomogeneities (Ω) have electroelastic moduli L_f , while the matrix has electroelastic moduli L_m . The composite is subjected to the far-field uniform applied stress and electric displacement Σ^o . As shown by

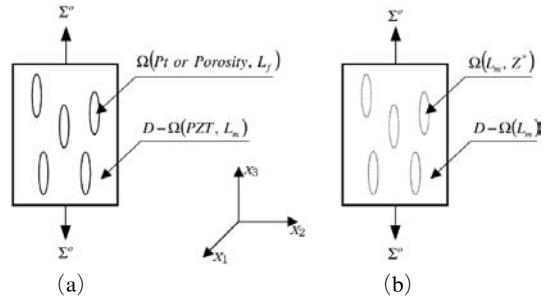


Fig. 1 An analytical model for computing the effective electroelastic properties of piezoelectric composite containing platinum or porosities, (a) original problem, which is converted to (b) Eshelby's equivalent inclusion problem

Eshelby (1957) in the elastic problems, the inhomogeneity in the piezoelectric solid can be simulated by an equivalent inclusion, resulting in Fig. 1(b).

When the composite is subjected to a far-field uniform load and electric displacement Σ^o , the stress and electric displacement in the inhomogeneity, Σ_f , can be written as :

$$\Sigma_f = L_f(Z^o + \bar{Z} + Z) = L_m(Z^o + \bar{Z} + Z - Z^*) \tag{4}$$

In Eq. (4), D is the disturbance of the uniform fields due to the presence of the inhomogeneities and \bar{Z} is the average disturbance of the strain and electric field in the matrix. Z^* is the fictitious eigenstrain and electric field by means of equivalent inclusion method (Eshelby, 1957). Z^o is the uniform strain and electric field by Σ^o that would exist in the absence of the inhomogeneity and related as

$$\Sigma^o = L_m Z^o \tag{5}$$

The stress and electric displacement in the matrix, Σ_m , can be written as :

$$\Sigma_m = L_m(Z^o + \bar{Z}) \tag{6}$$

The disturbed strain and electric field in the inclusion, Z , are related with fictitious eigenfields Z^* through piezoelectric Eshelby tensor S as :

$$Z = SZ^* \tag{7}$$

With the boundary condition Σ^o and the volume integral of the stress and electric displacement over the entire composite domain, the average

strain and electric field \bar{Z} are expressed as

$$\bar{Z} + v_f(Z - Z^*) = 0 \quad (8)$$

where v_f denotes volume fraction of porosity or metal. Z^* is computed from Eqs. (4), (5), (7), and (8) as

$$Z^* = -B(L_f - L_m)Z^o \quad (9)$$

where $B = \{(L_f - L_m)[(1 - v_f)S + v_f I] + L_m\}^{-1}$ and I is 9×9 identity matrix.

The overall strain and electric field of the composite Z_c can be obtained as the weighted volume average of that over each phase :

$$Z_c = (1 - v_f)Z_m + v_f Z_f = Z^o + v_f Z^* \quad (10)$$

where Z_m and Z_f represent total strain and electric fields in the matrix and filler, respectively.

The overall stress and electric displacement of the composite are given by

$$\Sigma_c = (1 - v_f)\Sigma_m + v_f \Sigma_f = \Sigma^o \quad (11)$$

The effective electroelastic moduli of the composite is defined as

$$\Sigma_c = L_c Z_c \quad (12)$$

From Eqs. (5), (9), and (10) ~ (12), the effective electroelastic moduli of the composite can be derived as

$$L_c = L_m [I - v_f B (L_f - L_m)]^{-1} \quad (13)$$

3. Results and Discussions

In this section, the effective electroelastic moduli of the porous and non-porous composites predicted by the present model are compared with the existing experimental results. The material properties of the piezoelectric matrix material

are tabulated in Table 1 (Li et al., 2003 ; Takagi et al., 2002), where data marked by * are not available from the literatures and are assumed. The model composite with porosities is PZT matrix containing porosities, while that of the non-porous composite is PZT matrix containing Pt. The piezoelectric matrix and Pt are considered as transversely isotropic and isotropic solids, respectively. Since the porosities were observed to be elongated along the perpendicular direction of an applied load during the die-pressing process with SEM micrographs (Li et al., 2003), the aspect ratio of porosities is reasonably assumed to be 1/10. The aspect ratio of Pt is assumed to be 1 due to both particle reinforced composite and SEM micrographs (Takaki et al., 2003) and its Young's modulus and Poisson's ratio are 146.9 GPa and 0.39, respectively.

The effective elastic constants of the porous and non-porous piezoelectric composites are computed as a function of volume fraction of both porosity and Pt and compared with the existing experimental results from the literatures (Li et al., 2003 ; Takaki et al., 2002). Since the experimental results for the effective elastic constants of the porous composite, C_{11} , C_{12} and C_{44} , are available, these data are compared with the predicted results by the present model. The assumption of C_{13} and C_{33} for computations in Table 1 would not influence the predicted results greatly. The predicted and experimental results for both the porous and non-porous composites are shown in Fig. 2. Fig. 2(a) shows that all the components of the elastic constants decrease monotonically with increasing porosity. These trends are consistent with the predicted results by Dunn et al. (1993³)

Table 1 Electroelastic material properties of piezoelectric matrix (Dunn and Taya, 1993c ; Li et al., 2003 ; Takaki et al., 2002)

	C [GPa]					e [C/m ²]			κ	
	C ₁₁	C ₁₂	C ₁₃	C ₃₃	C ₄₄	e ₃₁	e ₃₃	e ₁₅	κ ₁₁ /κ ₀ **	κ ₃₃ /κ ₀
PZT ^a	142	94.6	94.6*	128*	23.8	-5.17	20*	12.3*	916*	1900*
PZT ^b	146	95.4	94.3	128	25.3	-3.94	17.5	12.3*	916*	1654

PZT^a: material data for the porous composite

PZT^b: material data for the non-porous composite

* : not available

** : κ₀ = 8.85 × 10⁻²² [C²/Nm²] = permittivity of free space

and Wu (2000). However, the elastic constants of PZT/Pt composite increase with increasing Pt due to the use of stiffer fillers. The predictions for the composites by the present model are in good agreement with the experimental results.

As shown in Fig. 3, the effective piezoelectric constants of the porous and non-porous piezoelectric composites are computed and compared with experimental results (Li et al., 2001 ; 2003). For the prediction of the piezoelectric constants, d_{31} and d_{33} , of the porous composite, e_{33} and e_{15} of PZT are not available in the literature, so these are assumed to predict well piezoelectric constants d_{31} and d_{33} of the composite with $\nu_f=0$. These assumed values in Table 1 would be the same order as the piezoelectric constants of real

PZT material (Dunn and Taya, 1993c). For both the porous and non-porous composites, piezoelectric constants, d_{33} and d_{31} , are predicted to decrease in magnitude with increasing volume fraction of porosity or Pt. These trends are consistent with the predicted results by other researchers (Dunn and Taya, 1993c ; Wu, 2000). It can be concluded from Fig. 3 that the predicted results for both composites are in good agreement with the experimental results.

As shown in Fig. 4, the effective dielectric constants of the porous and non-porous piezoelectric composite are computed and compared with experimental results (Li et al., 2001 ; 2003). For predicting the dielectric constants of the porous,

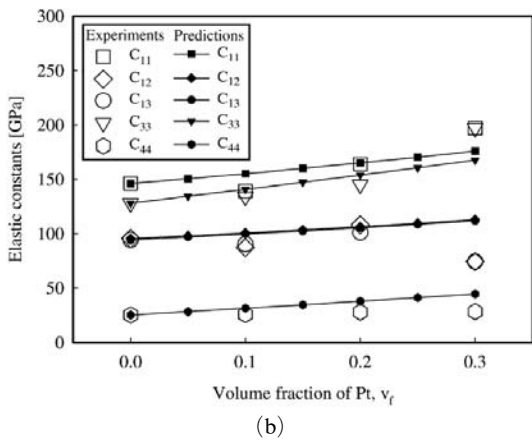
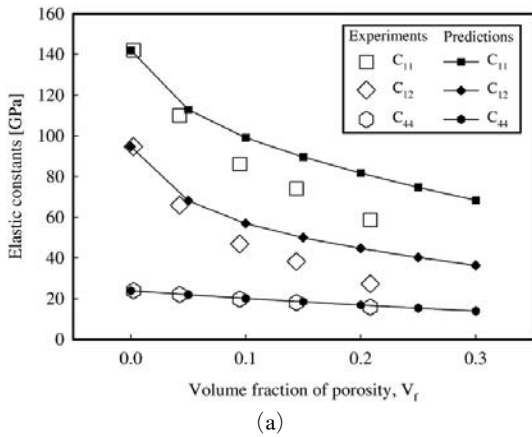


Fig. 2 Comparison of the predicted and experimental results for the effective elastic constants of the porous and non-porous composites : (a) porous composite, (b) non-porous composite

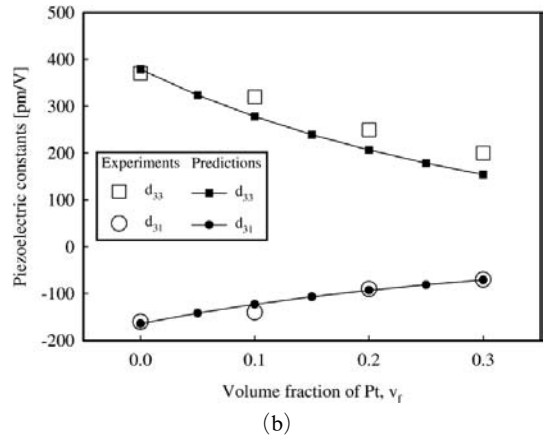
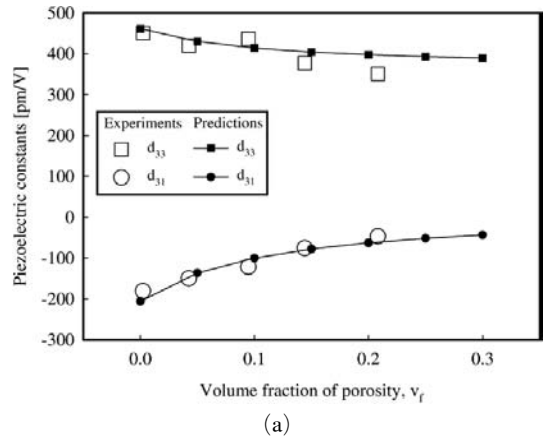


Fig. 3 Comparison of the predicted and experimental results for the effective piezoelectric constants of the porous and non-porous composites : (a) porous composite, (b) non-porous composite

dielectric constants κ_{33} and κ_{11} of PZT^a are assumed to predict well dielectric constant of the composite with $f=0$. The κ_{11} of PZT^b are assumed to be 916, whose effect on the dielectric constant is numerically confirmed to be negligible. In Fig. 4(a), the effective dielectric constant of the porous composite is seen to decrease rapidly with increasing porosity. The predictions are in good agreement with the experimental results.

In Fig. 4(b), the effective dielectric constants of the non-porous composite are predicted to decrease linearly with increasing volume fraction of Pt particles. However, the experimental results are shown to increase rapidly with increasing volume fraction of Pt particles. It has been ex-

perimentally observed that larger tensile thermal strain reduces the dielectric constant of the composite, while the larger compressive thermal strain increases the dielectric constant of the composite (Taylor et al., 2002). For the porous composite, the thermal mismatch strain between the porosity and matrix can be ignored, so the predicted dielectric constants are in good agreement with the experimental results. However, the thermal mismatch strain for the non-porous composite cannot be ignored, so the predictions of the dielectric constant are not in agreement with the experimental results. The coefficients of thermal expansion for PZT and Pt are 1.5×10^{-6} and 8.9×10^{-6} , respectively. According to Taylor et al. (2002), the compressive thermal mismatch strain in the piezoelectric matrix is generated during cooling process, so the dielectric constants of the composite increase with increasing volume fraction of Pt. It can be concluded that Eshelby type micromechanics model predicts well the effective electroelastic moduli of the porous and non-porous composite except the dielectric constants. Thus, thermal stress generated in the composite during processing has to be accounted for better prediction of the effective dielectric constants of the composite.

4. Conclusions

Eshelby type micromechanics model with a newly developed piezoelectric Eshelby tensor is proposed for predicting the effective electroelastic moduli of the composite. The model is applied for both the porous and non-porous piezoelectric composites. The prediction of the effective electroelastic moduli of the composites shows good agreement with the existing experimental results. However, predicted dielectric constant of the non-porous composite shows perfectly different trend from the experimental results. It can be concluded through the study that the Eshelby type model shows the limitation in predicting the dielectric constants of the non-porous piezoelectric composites. As shown by Taylor et al. (2002), the thermal stress generated in the composite during processing has to be accounted for better

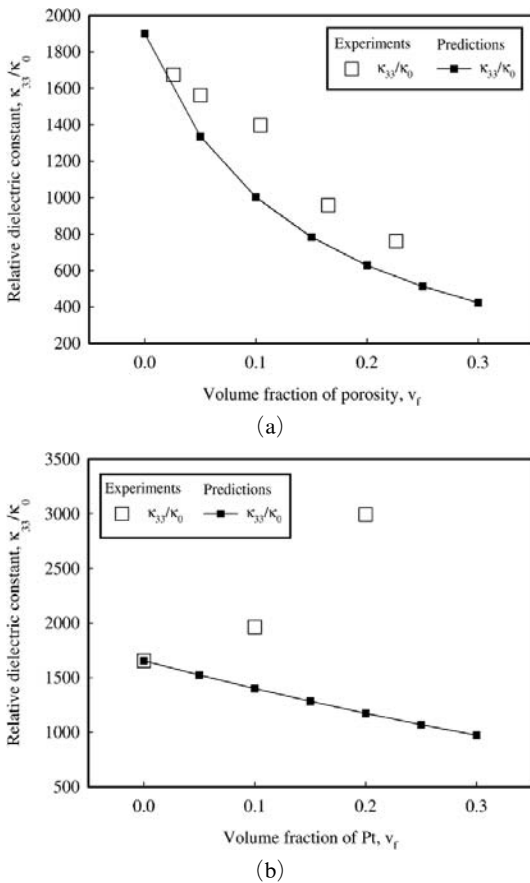


Fig. 4 Comparison of the predicted and experimental results for the effective dielectric constants of the porous and non-porous composites: (a) porous composite, (b) non-porous composite

prediction of the effective dielectric constants of the composite with the Eshelby type model.

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