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Effective Piezoelectric Properties of Composite Materials

Submitted by

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A thesis submitted in partial fulfilment of the requirements for the Degree of Master of Philosophy in Applied Physics

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Abstract

The Poon and Shin approach of finding an explicit formula for the effective dielectric constant of 0-3 composites was extended to obtain two explicit expressions for the prediction of the elastic properties (bulk modulus and shear modulus) of isotropic 0-3 composites. Predictions using these two expressions were compared with experimental data for elastic properties of a glass/epoxy composite. Good agreements, even for high volume fractions of the glass fibers were resulted. These two expressions were then combined with Poon and Shin's explicit effective dielectric formula into the calculation scheme of Wong et al. As a result, two explicit formulas for the prediction of d_{31} and d_{33} values for binary 0-3 piezoelectric composites were obtained. Comparisons of the predictions made by these explicit formulas, Wong et. al.'s scheme and the d_{31} of PZT/PVDF and d_{33} published experimental of data of PbTiO₃/P(VDF/TeFE) were presented.

Another pair of explicit formulae for the effective piezoelectric coefficients (d_{31} and d_{33}) of 0-3 composite of ferroelectric spheres embedded in a ferroelectric matrix taking into account the piezoelectric properties were also derived based

on Poon and Shin approach, By assuming that both phases were dielectrically and elastically isotropic even they were polarized, we were able to express the effective piezoelectric coefficients directly in terms of the properties of the constituents. Predictions made were then compared with published experimental data of the d_{31} of a PZT/PVDF composite (in which only the ceramic phase was polarized), the d_{33} of a PZT/P(VDF-TrFE) composites (with both phases polarized in the same direction) and d_{31} , d_{33} of a PZT/P(VDF-TrFE) composite (with the two phases polarized in opposite directions). Fairly good agreements were demonstrated. For the first two cases, results showed that both our model and Wong et. al.'s scheme had comparable performance. However, for the last case, our model gave more favourable predictions.

Effective piezoelectric coefficients of 1-3 piezoelectric fibre composites were also considered. Two explicit formulae for the effective piezoelectric stress coefficients (e_{31} and e_{33}) were derived based on an effective medium theory (EMT) method, under the assumptions that both phases were transversely isotropic and the electric field strengths inside the constituents were equal to the applied electric field. The results obtained were then combined with Chen model to evaluate the longitudinal piezoelectric strain coefficient d_{33} . Apart from the analytical EMT method, the effective piezoelectric coefficients of 1-3 composite were also calculated by a numerical EMT scheme. Results from both schemes were compared with the published experimental data of d_{33} of a 1-3 PZT/epoxy composite and the numerical values of e_{31} and e_{33} estimated by a finite element method of a 1-3 PZT/polymer composite.

List of Publications

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Chapter 1 Introduction

1.1 Background

Piezoelectricity was discovered by Pierre and his brother Jacques in 1880. They found that for some material, an electric field is induced inside when a mechanical stress is applied on it. This is called the direct piezoelectric effect. In 1881, they discovered the converse piezoelectric effect: some materials undergo deformation when electric field is applied. Most of these materials have a perovskite ABO₃ structure and have spontaneous polarization at room temperature. However, before poling, these piezoelectric materials do not exhibit any piezoelectric effect due to the random orientations of the polarization inside the domains in the materials. When a large electric field is applied to pole the material, the spontaneous polarization inside each domain will be reoriented along the electric field direction. As a result, permanent polarization exists and the material can exhibit piezoelectric effect. The piezoelectric coefficients d of a piezoelectric material are defined by the following equations

$$S_{ij} = d_{kij}E_k + s_{ijkl}^E T_{kl}$$
$$D_i = \varepsilon_{ik}^T E_k + d_{ikl}T_{kl}$$
(1.1)

where S, E, T, D, s and ε are strain, electric field, stress, electric displacement, elastic compliance and permittivity respectively. The superscripts E and T indicate that the elastic compliance and the permittivity are measured under constant electric field and constant applied stress respectively. The subscripts i, j, k, l can take values 1, 2 or 3, representing the X, Y and Z directions respectively.

To simplify the notation, the ordered pairs ij or kl can be replaced by a single index p or q as follows [ANSI/IEEE Std., 176-1987]

 $11 \rightarrow 1$, $22 \rightarrow 2$, $33 \rightarrow 3$, $23 \text{ or } 32 \rightarrow 4$, $31 \text{ or } 13 \rightarrow 5$, $12 \text{ or } 21 \rightarrow 6$.

Equations (1.1) can then be rewritten as

$$S_{p} = d_{kp}E_{k} + s_{pq}^{E}T_{q}$$
$$D_{i} = \varepsilon_{ik}^{T}E_{k} + d_{iq}T_{q}$$
(1.2)

Piezoelectric materials have been widely used in ultrasonic transducers, hydrophones and sensors [D. Stansfield, 1990]. Piezoceramics are usually good choices because of their high electromechanical coupling factor, which is defined as the square root of the ratio of output electrical (mechanical) energy to input mechanical (electrical) energy, and piezoelectric coefficients. However, piezoceramics have the limitations of having high stiffness constants and low hydrostatic piezoelectric coefficient $d_h \ (\equiv d_{31} + d_{32} + d_{33})$. Piezoelectric polymers such as polyvinylidene fluoride (PVDF) and its copolymer vinylidene fluoride /trifluorethylene (VDF/TrFE) are alternatives because they have high mechanical flexibility and hydrostatic piezoelectric coefficient. However, when compared with piezoceramics, piezoelectric polymers have much lower piezoelectric coefficients. Piezoelectric composites overcome the limitations of these single phase materials by taking advantages of each constituent and they can be tailor-made for specific applications. For a binary composite, there are ten possible connectivities of the constituent materials (0-0, 0-1, 0-2, 0-3, 1-1, 1-2, 1-3, 2-2, 2-3 and 3-3) [Newnham et al., 1978, Tressler et. al., 1999]. In this notation, the first (second) digit denotes the dimension of the inclusion phase (matrix phase). Among the ten connectivities, 0-3 and 1-3 are the two most important morphologies [Dias and Das-Gupta, 1994].

0-3 piezoelectric composites usually consist of piezoelectric particles embedded in a matrix that may or may not be piezoelectric. For 1-3 composites, piezoelectric rods or fibers are embedded instead. In this project, we will concentrate on finding the effective piezoelectric coefficients of 0-3 and 1-3 composites.

1.2 Literature review

In this section, we shall use the subscripts i and m to denote the inclusion phase and the matrix phase respectively. We shall use the subscripts 1, 2 and 3 to represent the X, Y and Z (the poling direction) directions respectively.

1.2.1 Literature review on 0-3 piezoelectric composites

Earlier modelling studies on 0-3 piezoelectric composites usually considered piezoelectric spherical or ellipsoidal inclusions in a non-piezoelectric medium. The researchers usually did not specify which piezoelectric constant were being considered. Furukawa et. al.'s [Furukawa et. al., 1976], Yamada et. al.'s [Yamada et. al., 1982], Jayasundere et. al.'s [Jayasundere et. al., 1994] and Prasad et. al.'s [Prasad et. al., 1996] models are some typical examples.

Furukawa et al. [Furukawa et. al., 1976] proposed a model of 0-3 composite materials that consisted of an inner sphere representing the inclusion phase and a concentric spherical shell representing the matrix phase. The combination was taken as a representative unit and it was surrounded by a homogeneous medium having the gross overall effective dielectric and elastic properties of the representative unit itself. They further assumed that both phases were isotropic and incompressible. Based on these assumptions, expressions of the effective dielectric constants (ε), elastic constants (c) and piezoelectric coefficients (d)

were derived as follows

$$\varepsilon = \frac{2\varepsilon_m + \varepsilon_i - 2\phi(\varepsilon_m - \varepsilon_i)}{2\varepsilon_m + \varepsilon_i + \phi(\varepsilon_m - \varepsilon_i)}\varepsilon_m$$
(1.3)

$$c = \frac{3c_m + 2c_i - 3\phi(c_m - c_i)}{3c_m + 2c_i + 2\phi(c_m - c_i)}c_m$$
(1.4)

$$d = \phi L_E L_T d_i \tag{1.5}$$

where ϕ is the volume fraction of the inclusion phase. L_E and L_T were called the local field coefficients and were defined as follows

$$L_E = \frac{E_i}{E} \tag{1.6}$$

$$L_T \equiv \frac{T_i}{T} \tag{1.7}$$

where E and T are the average electric field and average stress over a composite respectively.



Fig 1.1 An illustrated diagram for Furukawa's model

In 1979, Furukawa et. al. [Furukawa et. al., 1979] considered the contribution of the piezoelectric properties of the matrix phase. The effective piezoelectric coefficients of the composite was derived to be

$$d = \phi L_E L_T d_i + \frac{1}{1 - \phi} (1 - \phi L_E) (1 - \phi L_T) d_m$$
(1.8)

Yamada et al. [Yamada et al., 1982] studied the dielectric and the piezoelectric properties of a composite composed of piezoelectric ellipsoidal particles embedded in a dielectric matrix. The effective dielectric constant was found to

$$\varepsilon = \varepsilon_m \{ 1 + \frac{n\phi(\varepsilon_i - \varepsilon_m)}{n\varepsilon_m + (\varepsilon_i - \varepsilon_m)(1 - \phi)} \}$$
(1.9)

where n was called the shape parameter of the inclusion.

The effective piezoelectric coefficient of the composite was also found

$$d = \phi \alpha G d_i \tag{1.10}$$

where α was called the poling ratio and *G* was called the local electric field coefficient. Expressions of *G* and *n* can be found in Yamada et. al.'s paper [Yamada et. al., 1982].

Jayasundere [Jayasundere et. al., 1993, 1994] derived analytic expressions for the effective dielectric constant and an effective piezoelectric coefficient of 0-3 composites composed of piezoelectric spheres embedded in a dielectric matrix. For the dielectric problem, they considered the case when the composite was subjected to an external electric field so that each inclusion was polarized and can be represented by a dipole moment. Taking the interaction effects into account and using the condition $\varepsilon_i >> \varepsilon_m$, they derived an expression for the effective permittivity of the composite.

$$\varepsilon = \frac{\varepsilon_m (1-\phi) + \varepsilon_i \frac{3\phi\varepsilon_m}{\varepsilon_i + 2\varepsilon_m} \left[1 + 3\phi \frac{\varepsilon_i - \varepsilon_m}{\varepsilon_i + 2\varepsilon_m} \right]}{(1-\phi) + \frac{3\phi\varepsilon_m}{\varepsilon_i + 2\varepsilon_m} \left[1 + 3\phi \frac{\varepsilon_i - \varepsilon_m}{\varepsilon_i + 2\varepsilon_m} \right]}$$
(1.11)

For the piezoelectric problem, they considered the situation that the composite was subjected to an external stress, so that polarization was induced inside each inclusion due to the inclusion stress. Similar to the dielectric problem, assuming $\varepsilon_i \gg \varepsilon_m$ and c, $c_i \gg c_m$, the effective piezoelectric coefficient was derived to be

$$d \approx d_i \frac{\varepsilon}{\varepsilon_i} (1 + \frac{3\phi\varepsilon_i}{2\varepsilon_m + \varepsilon_i})$$
(1.12)

The effective permittivity ε appeared in equation (1.12) was given by equation (1.11).

Parasad et. al. [Prasad et. al., 1996] derived theoretically an effective piezoelectric stress coefficient based on Jayasundere [Jayasundere et. al., 1993] and Furukawa [Furukawa et. al., 1976] results. They found that the effective piezoelectric stress coefficient of the composite was as follows

$$e = \frac{1}{\phi} \frac{c_m - c_i}{c_m - c_i} \frac{\varepsilon_m - \varepsilon}{\varepsilon_m - \varepsilon_i} e_i$$
(1.13)

where the stiffness constant c and the dielectric constant ε were determined by equations (1.4) and (1.11) respectively.

All the above models were developed based on the assumption that the

piezoelectric activity of the composite was contributed by the inclusion phase only. Their results usually showed large derivations for available experimental data, especially at high volume fractions of the inclusions.

Recently, more researchers [Levin et. al., 2000, Levin and Luchaninov, 2001, Wong et. al., 2001, 2003] focused on the piezoelectric problem of 0-3 composites at high volume fractions of the inclusions.

For example, Levin et. al. [Levin et. al., 2000, Levin and Luchaninov, 2001] studied the effective constants for a pyroelectric 0-3 composite. The matrix phase and the inclusion phase considered were polarized in opposite directions. For high volume fractions of the inclusions, they used an effective field method, which is a kind of self-consistent schemes, to estimate the effective properties of the composites. The derived expressions of the piezoelectric stress coefficients (e_{31} , e_{33} and e_{15}) were

$$e_{31} = e_{31m} + \frac{\phi}{\Delta} [\alpha_1 e_{31i} - \frac{\alpha_2}{2} (C_{11i} + C_{12i}) - \alpha_3 C_{13i}]$$
(1.14)

$$e_{33} = e_{33m} + \frac{\phi}{\Delta} [\alpha_1 e_{33i} - \alpha_2 C_{13i} - \alpha_3 C_{33i}]$$
(1.15)

$$e_{15} = e_{15m} + \phi \frac{e_{15i}B_5 - 4C_{44i}Q_2}{B_5b_1 - 4q_2Q_2}$$
(1.16)

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where the expressions for α_k (k=1-3), Δ , Q_2 , B_5 , b_1 and q_2 can be found in their papers [Levin et. al., 2000, Levin and Luchaninov, 2001].

Wong et. al [Wong et. al., 2001] considered firstly the piezoelectric problem at low volume fractions and built their theoretical models based on the solutions of the dielectric [Wong et. al., 2001] and the mechanical [Goodier, 1933] problems of a single spherical inclusion composite. The effective piezoelectric coefficients were found to be

$$d_{31} = \phi L_E[(L_T^{\perp} + L_T^{\prime\prime})d_{31i} + L_T^{\perp}d_{33i}] + (1 - \phi)\overline{L}_E[(\overline{L}_T^{\perp} + \overline{L}_T^{\prime\prime})d_{31m} + \overline{L}_T^{\perp}d_{33m}] \quad (1.17)$$

$$d_{33} = \phi L_E [2L_T^{\perp} d_{31i} + L_T^{\prime\prime} d_{33i}] + (1 - \phi) \overline{L}_E [2\overline{L}_T^{\perp} d_{31m} + \overline{L}_T^{\prime\prime} d_{33m}]$$
(1.18)

where L_E and L_T were called the electrical field factors and the stress field factors respectively and were defined as follows

$$\begin{split} L_{E} &= \frac{3\varepsilon_{m}}{(1-\phi)\varepsilon_{i} + (2+\phi)\varepsilon_{m}} \\ \overline{L}_{E} &= \frac{1-\phi L_{E}}{1-\phi} \\ L_{T}^{\perp} &= \frac{I}{1-\phi(1-3I)} - \frac{J}{1-\phi(1-3J)} \\ L_{T}^{\prime\prime} &= \frac{I}{1-\phi(1-3I)} + \frac{2J}{1-\phi(1-3J)} \\ \overline{L}_{T}^{\perp} &= \frac{-\phi L_{T}^{\perp}}{1-\phi} \\ \overline{L}_{T}^{\prime\prime} &= \frac{1-\phi L_{T}^{\prime\prime}}{1-\phi} \end{split}$$

$$I = \frac{1 - v_m}{1 + v_m} \frac{(1 + v_i)\mu_i}{2(1 - 2v_i)\mu_m + (1 + v_i)\mu_i}$$
$$J = \frac{5(1 - v_m)\mu_i}{(7 - 5v_m)\mu_m + 2(4 - 5v_m)\mu_i}$$

where ν and μ are the Poisson's ratio and the shear modulus respectively.

For the concentrated inclusion cases, they replaced L_E and L_T by new electrical field factors F_E and stress field factors F_T . Two piezoelectric equations for non-dilute cases were found

$$d_{31} = \phi F_E[(F_T^{\perp} + F_T^{\prime\prime})d_{31i} + F_T^{\perp}d_{33i}] + (1 - \phi)\overline{F}_E[(\overline{F}_T^{\perp} + \overline{F}_T^{\prime\prime})d_{31m} + \overline{F}_T^{\perp}d_{33m}] \quad (1.19)$$

$$d_{33} = \phi F_E[2F_T^{\perp}d_{31i} + F_T^{\prime\prime}d_{33i}] + (1 - \phi)\overline{F}_E[2\overline{F}_T^{\perp}d_{31m} + \overline{F}_T^{\prime\prime}d_{33m}] \quad (1.20)$$

where

$$F_{E} = \frac{1}{\phi} \frac{\varepsilon - \varepsilon_{m}}{\varepsilon_{i} - \varepsilon_{m}}$$

$$\overline{F}_{E} = \frac{1}{1 - \phi} \frac{\varepsilon_{i} - \varepsilon}{\varepsilon_{i} - \varepsilon_{m}}$$

$$F_{T}^{\perp} = \frac{1}{\phi} \left(\frac{1}{3} \frac{\frac{1}{k} - \frac{1}{k_{m}}}{\frac{1}{k_{i}} - \frac{1}{k_{m}}} - \frac{1}{3} \frac{\frac{1}{\mu} - \frac{1}{\mu_{m}}}{\frac{1}{\mu_{i}} - \frac{1}{\mu_{m}}}\right)$$

$$F_{T}^{\prime\prime\prime} = \frac{1}{\phi} \left(\frac{1}{3} \frac{\frac{1}{k} - \frac{1}{k_{m}}}{\frac{1}{k_{i}} - \frac{1}{k_{m}}} + \frac{2}{3} \frac{\frac{1}{\mu} - \frac{1}{\mu_{m}}}{\frac{1}{\mu_{i}} - \frac{1}{\mu_{m}}}\right)$$

$$\overline{F}_{T}^{\perp} = \frac{1}{1 - \phi} \left(\frac{1}{3} \frac{\frac{1}{k_{i}} - \frac{1}{k_{m}}}{\frac{1}{k_{i}} - \frac{1}{k_{m}}} - \frac{1}{3} \frac{\frac{1}{\mu_{i}} - \frac{1}{\mu_{m}}}{\frac{1}{\mu_{m}} - \frac{1}{\mu_{m}}}\right)$$

$$\overline{F}_{T}^{''} = \frac{1}{1-\phi} \left(\frac{1}{3} \frac{\frac{1}{k_{i}} - \frac{1}{k}}{\frac{1}{k_{i}} - \frac{1}{k_{m}}} + \frac{2}{3} \frac{\frac{1}{\mu_{i}} - \frac{1}{\mu}}{\frac{1}{\mu_{i}} - \frac{1}{\mu_{m}}}\right)$$

In the above equations, the effective dielectric permittivity ε , the effective bulk modulus k and the effective shear modulus μ were then determined by Bruggeman [Bruggeman, 1935] and Hashin [Hashin, 1962].

In 2003, Wong et. al. [Wong et. al., 2003] derived another two explicit expressions of the effective piezoelectric stress coefficients, based on an effective medium theory (EMT). The EMT formulae of the effective stress coefficients e_h (under hydrostatic loading) and e_s (under shear loading) were given by

$$e_{h} = (1-\phi)\overline{F}_{E}\overline{L}_{S}^{h}(e_{hm}-e_{hi}) + \{1+(\varepsilon_{i}-\varepsilon)[\frac{\varepsilon_{i}+4\varepsilon_{m}}{(\varepsilon_{i}+2\varepsilon_{m})^{2}}\overline{L}_{S}^{h}-\frac{\varepsilon_{i}+4\varepsilon}{(\varepsilon_{i}+2\varepsilon)^{2}}] + \frac{(k_{i}-k)}{k_{i}}\times[\frac{(\varepsilon_{i}+8\varepsilon_{m})\varepsilon_{m}}{(\varepsilon_{i}+2\varepsilon_{m})^{2}}\overline{F}_{E}-\frac{(\varepsilon_{i}+8\varepsilon)\varepsilon}{(\varepsilon_{i}+2\varepsilon)^{2}}]\}e_{hi}$$
(1.21)

$$e_{s} = (1-\phi)\overline{F}_{E}\overline{L}_{s}^{s}(e_{sm}-e_{si}) + \{1+(\varepsilon_{i}-\varepsilon)[\frac{\varepsilon_{i}+4\varepsilon_{m}}{(\varepsilon_{i}+2\varepsilon_{m})^{2}}\overline{L}_{s}^{s}-\frac{\varepsilon_{i}+4\varepsilon}{(\varepsilon_{i}+2\varepsilon)^{2}}] + \frac{(\mu_{i}-\mu)}{\mu_{i}} \times [\frac{(\varepsilon_{i}+8\varepsilon_{m})\varepsilon_{m}}{(\varepsilon_{i}+2\varepsilon_{m})^{2}}\overline{F}_{E} - \frac{(\varepsilon_{i}+8\varepsilon)\varepsilon}{(\varepsilon_{i}+2\varepsilon)^{2}}]\}e_{si}$$

$$(1.22)$$

where

$$L_{S}^{h} = \frac{1}{\phi} \frac{k - k_{m}}{k_{i} - k_{m}}$$
$$L_{S}^{s} = \frac{1}{\phi} \frac{\mu - \mu_{m}}{\mu_{i} - \mu_{m}}$$
$$\overline{L}_{S}^{h} = \frac{1 - \phi L_{S}^{h}}{1 - \phi}$$
$$\overline{L}_{S}^{s} = \frac{1 - \phi L_{S}^{s}}{1 - \phi}$$

1.2.2 Literature review on 1-3 piezoelectric composites

This section gives a brief summary on some previous theoretical studies of the effective piezoelectric properties of 1-3 composites consist of piezoelectric inclusions embedded in a non-piezoelectric isotropic matrix.

Chan and Unsworth [Chan and Unsworth, 1989] and Smith [Smith, 1991, 1993] derived simple analytic expressions. They assumed that both phases were undergone the same strain in the Z directions, and the expressions of the effective piezoelectric coefficients were found to be

$$e_{31} = \frac{\phi e_{31i}(C_{11m} + C_{12m})}{\phi(C_{11m} + C_{12m}) + (1 - \phi)(C_{11i}^E + C_{12i}^E)}$$
(1.23)

$$e_{33} = \phi [e_{33i} - \frac{2\phi e_{31i}(C_{13i}^E - C_{12m})}{\phi(C_{11m} + C_{12m}) + (1 - \phi)(C_{11i}^E + C_{12i}^E)}]$$
(1.24)

$$d_{31} = \phi [d_{31i} - \frac{(1-\phi)d_{33i}(s_{13i} - s_{12m})}{\phi s_{11m} + (1-\phi)s_{33i}^E}]$$
(1.25)

$$d_{33} = d_{33i} \frac{\phi s_{11m}}{\phi s_{11m} + (1 - \phi) s_{33i}^E}$$
(1.26)

where $C_{\alpha\beta}$ and $s_{\alpha\beta}$ were stiffness constants and elastic compliances respectively. Their models did not consider the geometry of the inclusion and were derived under the assumption that the inclusions had small aspect ratio. Here, the aspect ratio χ was defined as the ratio of the diameter (or width) to the length of the inclusion. It was found to be a critical parameter affecting the effective properties of 1-3 composites and had been studied by many authors [Cao et. al., 1992; Zhang et. al., 1993; Nan and Jin, 1993; Nan, 1994; Sottos and Li, 1994; Li and Sottos, 1995, 1995].

Cao et. al. studied profile of the Z component of the displacement of each constituent and the effective hydrostatic piezoelectric coefficients of the composite. Their model consisted of a single piezoelectric fiber embedded in a passive matrix. They found that when an external stress T_3 was applied, the two phases had different displacement profiles, as expected, due to their different elastic and piezoelectric properties. For the piezoelectric problem, a

hydrostatic stress was applied, and they found that as stress was transferred from the matrix phase to the inclusion phase, the induced surface charge density was increased. The derived expression for the effective hydrostatic piezoelectric coefficient was

$$d_h = \phi(\gamma_h d_{33i} + 2d_{31i}) \tag{1.27}$$

where γ_h was called the stress amplification factor, given by

$$\gamma_h = 1 + \frac{\eta}{\eta_1} \tag{1.28}$$

where

$$\eta = (\ell/a)I_1(\rho_{ai})[I_1(\rho_{Rm})K_1(\rho_{am}) - I_1(\rho_{am})K_1(\rho_{Rm})][(1 - 2\sigma_m)/Y_m - (1 - 2\sigma_i)s_{33i}]$$

$$\eta_1 = \sqrt{(2s_{33i}/C_{44i})}I_0(\rho_{ai})[I_1(\rho_{Rm})K_1(\rho_{am}) - I_1(\rho_{am})K_1(\rho_{Rm})]$$

$$+ \sqrt{(2/Y_m\mu_m}I_1(\rho_{ai})[I_1(\rho_{Rm})K_0(\rho_{am}) + K_1(\rho_{Rm})I_0(\rho_{am})]$$

and ℓ , a, R, Y_m , μ_m , s_{33i} , C_{44i} are the length, radius of the inclusion, radius of the composite, Young's modulus of the matrix phase, shear modulus of the matrix phase, elastic compliance of the inclusion and the stiffness constant of the inclusion, respectively. I_0 , K_0 , I_1 and K_1 are the zerothand first-order modified Bessel functions. Expressions of σ_m , σ_i and ρ can be found in their paper [Cao et. al., 1992]. Zhang et. al. [Zhang et. al., 1993] further extended Cao et. al.'s model. They considered the influence of deformation of each PZT rod by its eight nearest neighbor rods. Their model was shown to be applicable up to $\phi \approx 0.2$.

Nan and Jin [Nan and Jin, 1993] developed a theoretical model for the effective properties of piezoelectric 1-3 composites based on a multi-scattering theory [Nan, 1993]. Their work was compared with the experimental data of piezoelectric coefficients of 1-3 PZT/Epoxy composites. The model took the approximation that $C^0 = C_m$ and $\varepsilon^0 = \varepsilon_m$, where C^0 and ε^0 were the stiffness constant and the permittivity tensors of a homogeneous comparison medium. This method was called the nonself-consistency (NSC). The expressions for the effective piezoelectric stress coefficients were found to be

$$e_{31} = \phi e_{31i} \frac{(K + m_m)}{(K_i + m_m)}$$
(1.29)

$$e_{33} = \phi e_{33i} - 2\phi e_{31i} \frac{(C_{13i} - C_{13})}{(K_i + m_m)}$$
(1.30)

$$e_{15} = \frac{\phi e_{15i} \varepsilon_m (m_m + 2C_{55})}{[(1+\phi)\varepsilon_m + (1-\phi)\varepsilon_{11i}](m_m + 2C_{55i})}$$
(1.31)

where $K \equiv C_{11} + C_{12}, m \equiv C_{11} - C_{12}$

and C_{11} , C_{12} , C_{13} and C_{55} were the stiffness constants of the composite.

As the concentration of the inclusion increases, the anisotropy of the composite become significant and the NCS scheme can no longer be applied. For the concentrated cases, Nan [Nan, 1994] took another approximation that $C^0 = C$ and $\varepsilon^0 = \varepsilon$, where *C* and ε were the stiffness constant and the permittivity tensors of the composite. This was called the self-consistent effective medium theory (SCEMT). SCEMT expressions of three effective piezoelectric stress coefficients were

$$\left\langle \frac{e_{31}}{K+m} - \frac{e_{31i}}{K_i + m} \right\rangle = 0$$
 (1.32)

$$\left\langle e_{33i} - e_{33} - \frac{2e_{31i}(C_{13i} - C_{13})}{K_i + m} \right\rangle = 0$$
 (1.33)

$$\left\langle \frac{e_{15}}{\varepsilon_{11i} + \varepsilon_{11}} - \frac{4e_{15i}C_{55}}{(C_{55i} + 3C_{55})(\varepsilon_{11i} + \varepsilon_{11})} \right\rangle = 0$$
(1.34)

where the bracket represented volume averages.

When compared with the experimental data of the hydrostatic voltage coefficient g_h of PZT/Epoxy composite for two aspect ratios ($\chi = 0.210$ and $\chi = 0.1$), their predicted values agreed well up to $\phi \sim 0.5$.

Li and Sottos [Li and Sottos, 1995, 1995, Sottos and Li, 1994] considered a composite system with a single piezoelectric fiber embedded in a

non-piezoelectric polymer matrix, and there was an interlayer between the inclusion phase and the matrix phase. They investigated the effects of aspect ratio, matrix stiffness and the Poisson ratio of the interlayer on the effective piezoelectric coefficients of the composite. Expressions of the displacements and stresses components in all three phases were derived. They assumed that the composite was subjected to a compressive hydrostatic pressure p. Using the proper boundary conditions and evaluating the average stresses and strains components in all three phases, the effective hydrostatic piezoelectric constant of the composite was found from the definition and the volumetric average of the electric displacement of the composite.

He and Lim [He and Lim, 2003] studied the effect of interfacial sliding on the effective piezoelectric coefficients of 1-3 composites. They considered the case in which the composite was subjected to a longitudinal shear stress τ_0 and at the same time, an external transverse electric field E_0 was applied. Using proper boundary conditions, the stresses, strains, electric fields and electric displacements of the constituents were derived. For larger volume fractions of piezoelectric inclusions, the Mori-Tanaka mean field approximation [Mori and Tanaka, 1973] was adopted. The overall electromechanical responses of the

composite can be characterized by the volume-averaged stress $\langle \sigma_{23} \rangle$, strain $\langle \gamma_{23} \rangle$, electric field $\langle E_2 \rangle$ and electric displacement $\langle D_2 \rangle$. They were given by

$$\langle \sigma_{23} \rangle = \tau_0; \quad \langle E_2 \rangle = E_0$$
 (1.35)

$$\left\langle \gamma_{23} \right\rangle = \varphi_1(t,\phi)\tau_0 + \varphi_2(t,\phi)\tau_0 \tag{1.36}$$

$$\langle D_2 \rangle = \psi_1(t,\phi)\tau_0 + \psi_2(t,\phi)\tau_0$$
 (1.37)

$$\langle E_2 \rangle = \phi \langle E_{2i} \rangle + (1 - \phi) \langle E_{2m} \rangle$$
 (1.38)

$$\langle \sigma_{23} \rangle = \phi \langle \sigma_{23i} \rangle + (1 - \phi) \langle \sigma_{23m} \rangle$$
 (1.39)

where t was time and expressions of φ_1 , φ_2 , ψ_1 and ψ_2 can be found in their paper [He and Lim, 2003].

And the required stress and electric field components were given by

$$\langle \sigma_{23p} \rangle = A_{1p}(t,\phi)\tau_0 + A_{2p}(t,\phi)E_0$$
 (1.40)

$$\langle E_{2p} \rangle = B_{1p}(t,\phi)\tau_0 + B_{2p}(t,\phi)E_0$$
 (1.41)

where *p* denoted *i* or *m* and expressions of A_{1p} , A_{2p} , B_{1p} and B_{2p} can be found in their paper [He and Lim, 2003].

Berger et. al. [Berger et. al., 2005] applied the finite element method (FEM) for calculating the effective properties of piezoelectric fiber composites. In order to

use FEM method, the composite material was represented by a periodical structure of representative volume elements (RVEs). Each RVE contained a piezoelectric fiber. They used this approach to find out the effective stiffness constants and the effective piezoelectric stress coefficients of a PZT/polymer composite.

Ren and Fan [Ren and Fan, 2006] investigated the effects of the oblique angles, volume fraction and the material properties of the constituents on the hydrostatic response of 1-3 composites. Oblique angle was defined as the orientation angle with respect to the poling axis of the fibers embedded in the matrix. They found that the total coupling factor k' ($\equiv \sqrt{k_{33}^2 + k_{15}^2}$) was highest when the oblique angle was around 27° and when the volume fraction of the inclusion was around 0.87.

Ray and Pradhan [Ray and Pradhan, 2006] studied the performance of lamina made of 1-3 piezoelectric composite material. They assumed that the composite was poled in the Z direction and the composite was in a plain strain state (i.e. $S_{22i} = S_{22m} = 0$). They assumed that both phases had the same vertical strain and the same lateral stress. Under these approximations, the derived expressions were as follows

$$e_{31} = e_{31i} \left[1 - \frac{(1-\phi)C_{11i}}{(1-\phi)C_{11i} + \phi C_{11m}} \right]$$
(1.42)

$$e_{33} = \phi [e_{33i} - e_{31i} \frac{(1 - \phi)(C_{13i} - C_{13m})}{(1 - \phi)C_{11i} + \phi C_{11m}}]$$
(1.43)

1.3 Scope of this study

The aim of this project is to study theoretically the effective piezoelectric coefficients of 0-3 and 1-3 composites. As mentioned in the introduction, these composites are important engineering materials and their piezoelectric properties can be tailored to suit specific applications.

The following is the structure of the thesis. Chapter 2 is divided into two parts. In the first part, we employ Poon and Shin approach [Poon and Shin, 2004] to derive two explicit formulae of the effective mechanical properties of 0-3 composites. These two formulae, together with the explicit expression of dielectric property reported by Poon and Shin are incorporated into Wong et. al.'s model [Wong et. al., 2001] to give two explicit formulae for the effective piezoelectric strain coefficients. Comparisons of the prediction of our model, Furukawa model [Furukawa, 1976], Jayasundere et. al.'s model [Jayasundere et. al., 1994] and some published experimental data are presented. In the second part, we employ the same approach to consider the piezoelectric problem of 0-3 composites, but from the very beginning. Expressions of the effective piezoelectric stress coefficients (e_{31} and e_{33}) and the effective stiffness constants (C_{11} , C_{12} , C_{13} and C_{33}) are derived. The predicted values of the

model are compared with experimental data of piezoelectric coefficients of 0-3 composites having three different polarization states: only the inclusion phase is polarized; both phases are polarized in the same direction and both phases are polarized in opposite directions. In chapter 3, we employ an effective medium theory (EMT) to find explicit expressions of piezoelectric stress coefficients for 1-3 composites at high volume fraction limit. The expressions are compared with some experimental data and simulated data from finite element method reported in the literature. Chapter 4 concludes what we have done so far and suggested some possible future works.

2.1 Introduction

Ferroelectrics composites have been studied for many years [Dias and Das-Gupta, 1996]. Their effective piezoelectric coefficients are important parameters in the design of piezoelectric devices because their values can be modified by varying the inclusion content in the composites. Recently, there have been many experimental works on the effective piezoelectric coefficients of 0-3 PZT/polymer composites [Chan et. al., 1994, 1995, Chan et. al., 1998, Chan et. al., 1999, Ng et. al., 2000]. It has also been demonstrated [Chan et. al., 1999] that the effective piezoelectric properties can be modified further by using different poling methods due to the different signs of the piezoelectric coefficients.

The aim of this chapter is to develop theoretical models for the predictions of the effective piezoelectric properties of 0-3 composites based on Poon and Shin approach [Poon and Shin, 2004]. The main idea of this approach is to take into account the interaction between the particulates. A brief review on this approach will be given in section 2.2.
In section 2.3, we employ Poon and Shin approach in treating the elastic problem of the 0-3 composite. Two new formulae for the effective stiffness constants are obtained and the values predicted are compared with the experimental data of Smith [Smith, 1976]. For the effective piezoelectric coefficients comparisons, predictions made by our scheme, Wong et. al.'s model [Wong et. al., 2001] and Furukawa et. al.'s model [Furukawa et. al., 1976] are compared with experimental results of d_{31} of PZT/PVDF composites [Furukawa, 1989]. For d_{33} , predicted values calculated by our scheme, Wong et. al.'s model and Jayasundere et. al. [Jayasundere et. al., 1994] are compared with the experimental data of a PbTiO₃/P(VDF/TeFE) system [Zou et. al., 1996].

In section 2.4, we used the same approach, but from the very beginning, to treat the piezoelectric problem of 0-3 composites. Assuming that both phases remain dielectrically and elastically isotropic even when they are polarized, expressions of the effective piezoelectric stress coefficients and the effective stiffness constants are derived. The results obtained are then used to find the effective piezoelectric strain coefficients. Predictions made by our model and Wong et. al.'s model were compared with the experimental data of PZT/polymer composites having three different polarization states. Namely, only the inclusion phase is polarized; both phases are polarized in the same direction and the two phases are polarized in opposite directions.

In the following sections of this chapter, we shall use the symbols i and m to refer to the inclusion phase and the matrix phase respectively and use p to represent either i or m. We use subscripts 1, 2 and 3 to denote the X, Y and Z directions, respectively and use $\langle x \rangle$ to denote the volumetric average of the physical quantity x over the respective material.

2.2 A brief review of Poon and Shin approach

Poon and Shin [Poon and Shin, 2004] considered the dielectric problem of a single dielectric sphere with dielectric constant ε_i embedded in an infinite matrix with dielectric constant ε_m . When an external electric field is applied along the Z direction and suppose $\langle E_{3m} \rangle$ is the electric field in the matrix region far away from the inclusion, it can be shown that the electric field $\langle E_{3i} \rangle$ inside the inclusion, is uniform and parallel to $\langle E_{3m} \rangle$ [Wong et. al., 2001]. The relationship between the electric fields $\langle E_{3i} \rangle$ and $\langle E_{3m} \rangle$, and the corresponding electric displacements $\langle D_{3i} \rangle$ and $\langle D_{3m} \rangle$ is given by [Wong et.

al., 2001]

$$\langle D_{3i} \rangle - \langle D_{3m} \rangle = -2\varepsilon_m (\langle E_{3i} \rangle - \langle E_{3m} \rangle)$$
 (2.1)

Defining $\Delta D \equiv \langle D_{3i} \rangle - \langle D_{3m} \rangle$ and $\Delta E \equiv \langle E_{3i} \rangle - \langle E_{3m} \rangle$, it can be written in the form:

$$\Delta D = -2\varepsilon_m \Delta E \tag{2.2}$$

Equation (2.1) (or (2.2)) is valid for dilute suspension of the inclusions only. For finite volume fraction of the inclusions, the interaction effects between the inclusions become significant. Suppose we have two 0-3 composites with $\phi > 0$ and $\phi \to 0$ as shown in Fig. 2.1, in which the circle denotes an inclusion particle having dielectric constant ε_i and the shaded region denotes the matrix phase having the dielectric constant ε_m .



Figure 2.1 Composite with the volume fraction of inclusion (a) $\phi > 0$ and with (b) $\phi \rightarrow 0$ subjected to an external electric field E_3

Let δD_m be the electric displacement contributed by the inclusion phase,

$$\delta D_m \equiv \langle D_3 \rangle - \varepsilon_m \langle E_3 \rangle \tag{2.3}$$

where $\langle D_3 \rangle$ is the volumetric averaged electric displacement of the composite.

After simplifying equation (2.3), they got

$$\delta D_m = \phi(\varepsilon_i - \varepsilon_m) \langle E_{3i} \rangle \tag{2.4}$$

Poon and Shin [Poon and Shin, 2004] proposed that the electric displacement $\langle D_{3m} \rangle$ for the higher volume fractions could be approximated to the sum of two parts: one due to the pure medium and the other due to the total polarization of other inclusions δD_m . Equation (2.4) was modified to the following:

$$\Delta D = -2\varepsilon_m \Delta E + \phi(\varepsilon_i - \varepsilon_m) \langle E_{3i} \rangle$$
(2.5)

The effective dielectric constant ε of the composite is defined by

$$\langle D_3 \rangle = \varepsilon \langle E_3 \rangle$$
 (2.6)

Its explicit formulas was shown to be

$$\varepsilon = \varepsilon_m + \phi(\varepsilon_i - \varepsilon_m) \frac{1}{\phi + (1 - \phi) \frac{\varepsilon_i + 2\varepsilon_m - \phi(\varepsilon_i - \varepsilon_m)}{3\varepsilon_m}}$$
(2.7)

2.3 Explicit formulae of the effective stiffness constants

2.3.1 Theory

For elastic properties, we can, by analogy, replace the electric displacement D by the stress T and the electric field E by the strain S.

Consider a composite with a single sphere having bulk modulus k_i and shear modulus μ_i in an isotropic matrix having bulk modulus k_m and shear modulus μ_m subjecting to a uniform external stress T_3 . Suppose a uniform tension T_0 is acting in the matrix far away from the inclusion, Goodier [Goodier, 1933] has worked out, in spherical coordinates, the analytical solutions for the displacements and the stresses inside the inclusion and the matrix. By transforming Goodier's solution to Cartesian coordinates, we find that the stress and the strain inside the constituents are [Detailed derivation is given in appendix A],

$$T_{11i} = T_{22i} = 3k_i B_1 - 4\mu_i B_2, \qquad T_{33i} = 3k_i B_1 + 8\mu_i B_2$$
$$S_{11i} = S_{22i} = B_1 - 2B_2, \qquad S_{33i} = B_1 + 4B_2 \qquad (2.8)$$

where B_1 and B_2 are defined by

$$B_{1} \equiv \frac{3k_{m} + 4\mu_{m}}{9k_{m}(3k_{i} + 4\mu_{m})}T_{0}, \quad B_{2} \equiv \frac{5}{12}\frac{3k_{m} + 4\mu_{m}}{\mu_{m}(9k_{m} + 8\mu_{m}) + 6(k_{m} + 2\mu_{m})\mu_{i}}T_{0}$$

and we have

$$T_{11m} = T_{22m} = 0, T_{33m} = T_0$$

$$S_{11m} = S_{22m} = \frac{\frac{2\mu_m}{3} - k_m}{6k_m \mu_m} T_0, S_{33m} = \frac{2(k_m + \frac{\mu_m}{3})}{6k_m \mu_m} T_0 (2.9)$$

Using equations (2.8) and (2.9), we get

$$\delta T_1 = A \delta S_1 + B \delta S_2 + B \delta S_3$$

$$\delta T_2 = B \delta S_1 + A \delta S_2 + B \delta S_3$$

$$\delta T_3 = B \delta S_1 + B \delta S_2 + A \delta S_3$$
(2.10)

where
$$\delta T_j \equiv T_{jji} - T_{jjm}$$
, $\delta S_j \equiv S_{jji} - S_{jjm}$ (j = 1,2,3) (2.11)

and
$$A = \frac{10}{9}\mu_m (-3 + \frac{2\mu_m}{k_m + 2\mu_m}), \quad B = \frac{1}{9}\mu_m (-3 - \frac{10\mu_m}{k_m + 2\mu_m})$$
 (2.12)

Similar to the electrical case, equation (2.10) is not a good approximation when there are several inclusions inside the matrix. Consider two composites with $\phi > 0$ (shown in Fig. 2.2. (a)) and $\phi \rightarrow 0$ (shown in Fig. 2.2 (b)) having strains $\langle S_{11} \rangle$, $\langle S_{22} \rangle$ and $\langle S_{33} \rangle$ along X, Y and Z directions.



Figure 2.2 (a) A Composite with $\phi > 0$ was undergone the strains $\langle S_{11} \rangle$,

 $\langle S_{22} \rangle$ and $\langle S_{33} \rangle$ along X , Y and Z directions respectively.



Figure 2.2 (b) A composite with $\phi \to 0$ was undergone the strains $\langle S_{11} \rangle$, $\langle S_{22} \rangle$ and $\langle S_{33} \rangle$ along X, Y and Z directions respectively.

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In Fig. 2.2 (a), the spheres represent the inclusion particles with bulk modulus k_i and shear modulus μ_i . The shaded region represents the matrix phase having bulk modulus k_m and shear modulus μ_m . For non-dilute suspension of the inclusions, we need to consider the interaction effects between the inclusions. Suppose $\langle T_{11} \rangle$ and $\langle T_{110} \rangle$ are the stresses of the composites along X direction with $\phi > 0$ and $\phi \rightarrow 0$ respectively. δT_{m1} is contributed by interaction between the inclusions which is defined by

$$\delta T_{m1} \equiv \left\langle T_{11} \right\rangle - \left\langle T_{110} \right\rangle \tag{2.13}$$

Since the 0-3 composite, as a whole, is an isotropic material, its elastic behaviour can be described by just two coefficients, for example, the effective bulk modulus k and the effective shear modulus μ , defined by the overall stress-strain relationship,

$$\begin{pmatrix} \langle T_{11} \rangle \\ \langle T_{22} \rangle \\ \langle T_{33} \rangle \end{pmatrix} = \begin{pmatrix} E & F & F \\ F & E & F \\ F & F & E \end{pmatrix} \begin{pmatrix} \langle S_{11} \rangle \\ \langle S_{22} \rangle \\ \langle S_{33} \rangle \end{pmatrix}$$
(2.14)

where

$$E \equiv k + \frac{4}{3}\mu, \quad F \equiv k - \frac{2}{3}\mu$$

Using equations (2.13) and (2.14), $\langle T_{110} \rangle$ can be expressed in terms of the strains. Equation (2.13) can be expressed as follows

$$\delta T_{m1} = \frac{1}{V} \int_{V} \{ T_{11} - [(k_m + \frac{4}{3}\mu_m)S_{11} + (k_m - \frac{2\mu_m}{3})S_{22} + (k_m - \frac{2\mu_m}{3})S_{33}] \} dV$$
(2.15)

where V is the volume of the composite.

After simplifying, we get

$$\delta T_{m1} = \frac{1}{V} \int_{V_i} \left[(\Delta k + \frac{4}{3} \Delta \mu) S_{11i} + (\Delta k - \frac{2}{3} \Delta \mu) S_{22i} + (\Delta k - \frac{2}{3} \Delta \mu) S_{33i} \right] dV$$

$$\equiv \phi \left[(\Delta k + \frac{4}{3} \Delta \mu) \left\langle S_{11i} \right\rangle + (\Delta k - \frac{2}{3} \Delta \mu) \left\langle S_{22i} \right\rangle + (\Delta k - \frac{2}{3} \Delta \mu) \left\langle S_{33i} \right\rangle \right]$$

(2.16)

where $\Delta k \equiv k_i - k_m$, $\Delta \mu \equiv \mu_i - \mu_m$

Similarly, δT_{m2} and δT_{m3} are given by

$$\delta T_{m2} \equiv \frac{1}{V} \int_{V} \{ T_{22} - [(k_m - \frac{2}{3}\mu_m)S_{11} + (k_m + \frac{4\mu_m}{3})S_{22} + (k_m - \frac{2\mu_m}{3})S_{33}] \} dV$$

$$\equiv \phi [(\Delta k - \frac{2}{3}\Delta\mu) \langle S_{11i} \rangle + (\Delta k + \frac{4}{3}\Delta\mu) \langle S_{22i} \rangle + (\Delta k - \frac{2}{3}\Delta\mu) \langle S_{33i} \rangle]$$

(2.17)

$$\delta T_{m3} = \frac{1}{V} \int_{V} \{ T_{33} - [(k_m - \frac{2}{3}\mu_m)S_{11} + (k_m - \frac{2\mu_m}{3})S_{22} + (k_m + \frac{4\mu_m}{3})S_{33}] \} dV$$

$$= \phi [(\Delta k - \frac{2}{3}\Delta\mu) \langle S_{11i} \rangle + (\Delta k - \frac{2}{3}\Delta\mu) \langle S_{22i} \rangle + (\Delta k + \frac{4}{3}\Delta\mu) \langle S_{33i} \rangle]$$

(2.18)

By analogy, we add an additional term to equation (2.10) and rewrite it in a

matrix form

$$\begin{pmatrix} \Delta T_1 \\ \Delta T_2 \\ \Delta T_3 \end{pmatrix} = \begin{pmatrix} A & B & B \\ B & A & B \\ B & B & A \end{pmatrix} \begin{pmatrix} \Delta S_1 \\ \Delta S_2 \\ \Delta S_3 \end{pmatrix} + \phi \begin{pmatrix} C & G & G \\ G & C & G \\ G & G & C \end{pmatrix} \begin{pmatrix} \langle S_{11i} \rangle \\ \langle S_{22i} \rangle \\ \langle S_{33i} \rangle \end{pmatrix}$$
(2.19)

where $\Delta T_{j} \equiv \langle T_{jji} \rangle - \langle T_{jjm} \rangle$, $\Delta S_{j} \equiv \langle S_{jji} \rangle - \langle S_{jjm} \rangle$ (j = 1, 2, 3) $C \equiv \Delta k + \frac{4}{3} \Delta \mu$, $G \equiv \Delta k - \frac{2}{3} \Delta \mu$ (2.20)

Using equations (2.12), (2.14) and (2.16) to (2.20), and the following relationships of the volumetric averages [Wong et. al., 2001],

$$\langle T_{jj} \rangle = \phi \langle T_{jji} \rangle + (1 - \phi) \langle T_{jjm} \rangle$$
 (2.21)

$$\langle S_{jj} \rangle = \phi \langle S_{jji} \rangle + (1 - \phi) \langle S_{jjm} \rangle$$
 (2.22)

the effective bulk modulus k and the effective shear modulus μ of the composite can be found [detailed derivation is given in Appendix B]:

$$k = k_m + \frac{\phi(k_i - k_m)(k_m + \frac{4}{3}\mu_m)}{(1 - \phi)[k_m + \frac{4}{3}\mu_m + (1 - \phi)(k_i - k_m)] + \phi(k_m + \frac{4}{3}\mu_m)}$$
(2.23)

$$\mu = \mu_m + \frac{\phi(\mu_i - \mu_m) \frac{5\mu_m (3k_m + 4\mu_m)}{6(k_m + 2\mu_m)}}{(1 - \phi)[\frac{5\mu_m (3k_m + 4\mu_m)}{6(k_m + 2\mu_m)} + (1 - \phi)(\mu_i - \mu_m)] + \phi \frac{5\mu_m (3k_m + 4\mu_m)}{6(k_m + 2\mu_m)}}{(2.24)}$$

2.3.2 Effective piezoelectric coefficients using the new stress field factors

Wong et al. [Wong et. al., 2001] have given explicit formulas for the effective piezoelectric d_{31} and d_{33} coefficients of 0-3 composites:

$$d_{31} = \phi F_E[(F_T^{\perp} + F_T'')d_{31i} + F_T^{\perp}d_{33i}] + (1 - \phi)\overline{F}_E[(\overline{F}_T^{\perp} + \overline{F}_T'')d_{31m} + \overline{F}_T^{\perp}d_{33m}]$$
(2.25)

$$d_{33} = \phi F_E [2F_T^{\perp} d_{31i} + F_T^{\prime\prime} d_{33i}] + (1 - \phi)\overline{F}_E [2\overline{F}_T^{\perp} d_{31m} + \overline{F}_T^{\prime\prime} d_{33m}]$$
(2.26)

where F_E and F_T are called the electric and stress field factors. Their definitions have been given in section 1.2.1.

For the dielectric constant ε appeared in the electric field factor, they used the Bruggeman formula [Bruggeman, 1935]

$$\frac{\varepsilon_i - \varepsilon}{\varepsilon_i^{\frac{1}{3}}} = (1 - \phi) \frac{\varepsilon_i - \varepsilon_m}{\varepsilon_m^{\frac{1}{3}}}$$
(2.27)

For the bulk k and shear μ modulus in the stress field factors, they used the Hashin model [Hashin, 1962].

Using their scheme, but employing equation (2.7) for the effective dielectric constant and the new formulas (equations (2.23) and (2.24)) for the effective elastic coefficients, we obtained two new explicit equations for the effective piezoelectric coefficients d_{31} and d_{33} .

$$d_{31} = \phi P_E[(P_T^{\perp} + P_T^{\prime\prime})d_{31i} + P_T^{\perp}d_{33i}] + (1 - \phi)\overline{P}_E[(\overline{P}_T^{\perp} + \overline{P}_T^{\prime\prime})d_{31m} + \overline{P}_T^{\perp}d_{33m}]$$
(2.28)

$$d_{33} = \phi P_E[2P_T^{\perp} d_{31i} + P_T^{\prime\prime} d_{33i}] + (1 - \phi)\overline{P}_E[2\overline{P}_T^{\perp} d_{31m} + \overline{P}_T^{\prime\prime} d_{33m}]$$
(2.29)

where

$$P_{E} = \frac{3\varepsilon_{m}}{3\varepsilon_{m}\phi + (1-\phi)[\varepsilon_{i} + 2\varepsilon_{m} - \phi(\varepsilon_{i} - \varepsilon_{m})]}, \quad \overline{P}_{E} = \frac{1-\phi P_{E}}{1-\phi}$$

$$P_{T}^{\perp} = \frac{1}{3} \left[\frac{L_{k}k_{i}}{k_{m} + \phi(k_{i} - k_{m})L_{k}} - \frac{L_{\mu}\mu_{i}}{\mu_{m} + \phi(\mu_{i} - \mu_{m})L_{\mu}} \right], \quad \overline{P}_{T}^{\perp} = \frac{-\phi P_{T}^{\perp}}{1-\phi}$$

$$P_{T}^{\prime\prime\prime} = \frac{1}{3} \left[\frac{L_{k}k_{i}}{k_{m} + \phi(k_{i} - k_{m})L_{k}} + \frac{2L_{\mu}\mu_{i}}{\mu_{m} + \phi(\mu_{i} - \mu_{m})L_{\mu}} \right], \quad \overline{P}_{T}^{\prime\prime} = \frac{1-\phi P_{T}^{\prime\prime}}{1-\phi}$$

and

$$L_{k} = \frac{(k_{m} + \frac{4}{3}\mu_{m})}{(1-\phi)[k_{m} + \frac{4}{3}\mu_{m} + (1-\phi)(k_{i} - k_{m})] + \phi(k_{m} + \frac{4}{3}\mu_{m})}$$
$$L_{\mu} = \frac{\frac{5\mu_{m}(3k_{m} + 4\mu_{m})}{6(k_{m} + 2\mu_{m})}}{(1-\phi)[\frac{5\mu_{m}(3k_{m} + 4\mu_{m})}{6(k_{m} + 2\mu_{m})} + (1-\phi)(\mu_{i} - \mu_{m})] + \phi\frac{5\mu_{m}(3k_{m} + 4\mu_{m})}{6(k_{m} + 2\mu_{m})}}$$

2.3.3 Comparisons with experimental data and discussions

In this subsection, we compare the predictions of the effective bulk modulus and the effective shear modulus using the two formulas (2.23) and (2.24) with the experimental data given by Smith [Smith, 1976]. The composite considered is a matrix of epoxy embedded with glass spheres. The Poisson's ratios of the glass (v_i) and the epoxy (v_m) are 0.23 and 0.394 respectively and the Young's modulus of the glass (Y_i) and the epoxy (Y_m) are 76.0 GPa and 3.01 GPa respectively. For an isotropic material, the bulk modulus and the shear modulus can be expressed in terms of Young's modulus and Poisson's ratio [Sadda, 1993].

$$k = \frac{Y}{3(1 - 2\nu)}$$
(2.30)

$$\mu = \frac{Y}{2(1+\nu)} \tag{2.31}$$

Fig. 2.3 shows the comparison results for the effective bulk modulus, in which we have plotted predictions based on our model and Hashin model [Hashin, 1962]. The bulk modulus of Hashin's model is given by

$$k = k_m + \frac{\phi(k_i - k_m)}{1 + (1 - \phi)\frac{k_i - k_m}{k_m + \frac{4}{3}\mu_m}}$$
(2.32)

Fig 2.3 shows that, at low volume fractions of the glass spheres, both Hashin's model and our model give good agreement with the experimental data. However, at higher volume fractions, Hashin model underestimates the effective bulk modulus while our model still fits relatively well to the experimental data.



Figure 2.3 Comparison of the effective bulk modulus predicted by this work (Equ.(2.23)) and Hashin's model (Equ.(2.32)) with experimental data of Smith [Smith, 1976]

Fig. 2.4 shows the comparison for the effective shear modulus. In this case, Hashin's model can only give the lower bound μ_l and the upper bound μ_u , as follows:

$$\mu_{l} = \mu_{m} (1 + \frac{15(1 - \nu_{m})(\frac{\mu_{i}}{\mu_{m}} - 1)\phi}{7 - 5\nu_{m} + 2(4 - 5\nu_{m})(\frac{\mu_{i}}{\mu_{m}} - (\frac{\mu_{i}}{\mu_{m}} - 1)\phi)})$$
(2.33)
$$\mu_{u} = \mu_{m} [1 + (\frac{\mu_{i}}{\mu_{m}} - 1)\frac{B_{1}}{A_{1} + B_{1}C_{1}}\phi]$$
(2.34)

where

$$A_{1} = \frac{42}{5\mu_{m}} \frac{\mu_{m} - \mu_{i}}{1 - \nu_{m}} \phi(\phi^{\frac{2}{3}} - 1)^{2} \vartheta$$

$$B_{1} = [(7 - 10\nu_{i}) - (7 - 10\nu_{m})\vartheta] 4\phi^{\frac{7}{3}} + (7 - 10\nu_{m})\vartheta$$

$$C_{1} = \frac{\mu_{i}}{\mu_{m}} + \frac{7 - 5\nu_{m}}{15(1 - \nu_{m})} (1 - \frac{\mu_{i}}{\mu_{m}}) + \frac{2(4 - 5\nu_{m})}{15(1 - \nu_{m})} (1 - \frac{\mu_{i}}{\mu_{m}})\phi$$
an
$$\vartheta = \frac{(7 + 5\nu_{i})\mu_{i} + 4(7 - 10\nu_{i})\mu_{m}}{35(1 - \nu_{m})\mu_{m}} \qquad (2.35)$$

It can be seen that, at low volume fractions, both the bounds of Hashin's model and our model can give reasonable predicted values, compared with the experimental data. However, when the volume fraction of the inclusions increases, the lower bound of Hashin's model fails to give reasonable predictions, while its upper bound values and predicted values from our model still show good agreement with the experimental data.



Figure 2.4 Comparison of the effective shear modulus predicted by this work (Equ.(2.24)), lower bound of shear modulus of Hashin model, μ_1 (Equ. (2.33)) and upper bound of shear modulus of Hashin model, μ_u (Equ. (2.34)) with experimental data of Smith [Smith, 1976].

For effective piezoelectric coefficients comparisons, predicted values using equations (2.28) and (2.29), Wong et. al.'s scheme [Wong et. al., 2001] and other models [Furukawa et. al., 1976, Jayasundere et. al., 1994] are compared with the experimental data given by Furukawa [Furukawa, 1989] and Zou et al. [Zou et. al., 1996], as shown in figures 2.5 and 2.6.

Fig. 2.5 shows the d_{31} comparison results for a PZT/PVDF system [Furukawa, 1989]. The Poisson's ratios of the PZT inclusion and the matrix are 0.3 and 0.4 respectively and the Young's modulus of the inclusion and the matrix are 58.7

GPa and 2.52 GPa respectively. The dielectric constants are 1900 for the inclusion and 14 for the matrix. The d_{31} and d_{33} values for the inclusion are -180 pC/N and 450 pC/N respectively. For this composite system, the matrix phase PVDF is not polarized and does not exhibit piezoelectric effect. This means that the effective piezoelectric activity of the composite is contributed by the inclusion phase only. For this comparison, Furukawa model [Furukawa et. al., 1976] is also included. Their expression for and d is given by

$$d = \phi L_E L_T d_i \tag{2.36}$$

where L_E and L_T are

$$L_E = \frac{3\varepsilon_m}{(1-\phi)\varepsilon_i + (2+\phi)\varepsilon_m}$$
(2.37)

$$L_T = \frac{5c_i}{3c_m + 2c_i - 3\phi(c_m - c_i)}$$
(2.38)

and c is the Young's modulus (or shear modulus) since both phases are assumed to be incompressible.

Fig. 2.5 shows that, for dilute suspension cases, our scheme and the scheme of Wong et al. using μ_u show similar performance, while the Furukawa model underestimates the piezoelectric coefficient. At higher volume fractions, the Furukawa model fails obviously, while our scheme and Wong et. al.'s scheme still show similar performance.



Figure 2.5 Predictions of the effective piezoelectric coefficient d₃₁ by this work (Equ.(2.28)), Wong et al.'s model [Wong et. al., 2001] using lower and upper bounds of shear modulus of Hashin [Hashin, 1962] and Furukawa's model [Furukawa et. al., 1976]. Experimental data are taken from [Furukawa, 1989] of Furukawa.

Fig. 2.6 shows the d_{33} comparison results from various models with the experimental data for a PbTiO₃/P(VDF/TeFE) system [Zou et. al., 1996]. They obtained their experimental values only at high volume fractions of the inclusions. The Poisson's ratios of the inclusion and the matrix are 0.22 and 0.4 respectively and the Young's modulus of the inclusion and the matrix are 126.7 GPa and 2.81 GPa respectively. The d_{31} and d_{33} values for the inclusion are -9.5 pC/N and 94 pC/N respectively. The dielectric constants are 150 for the inclusion and 6 for the matrix respectively. In their experiment, only the inclusion phase is polarized.

Jayasundere model [Jayasundere et. al., 1994] is also included for comparison.

Their expression for d is given below

$$d = d_i \frac{\varepsilon}{\varepsilon_i} \left(1 + \frac{3\phi\varepsilon_i}{2\varepsilon_m + \varepsilon_i}\right)$$
(2.39)

In equation (2.39), the effective permittivity ε they used is given by Jayasundere [Jayasundere et. al., 1993]:

$$\varepsilon = \frac{\varepsilon_m (1-\phi) + \varepsilon_i \frac{3\phi\varepsilon_m}{\varepsilon_i + 2\varepsilon_m} \left[1 + 3\phi \frac{\varepsilon_i - \varepsilon_m}{\varepsilon_i + 2\varepsilon_m} \right]}{(1-\phi) + \frac{3\phi\varepsilon_m}{\varepsilon_i + 2\varepsilon_m} \left[1 + 3\phi \frac{\varepsilon_i - \varepsilon_m}{\varepsilon_i + 2\varepsilon_m} \right]}$$
(2.40)

Fig. 2.6 shows that, for this case, almost all experimental data fall within the bounds of Wong et al. [Wong et. al., 2001]. Our scheme fits reasonably good to the data. On the other hand, the Jayasundere model obviously overestimates the coefficient.



Figure 2.6 Predictions of the effective piezoelectric coefficient d₃₃ by this work (Equ.(2.29)), Wong et al.'s model [Wong et. al., 2001] using lower and upper bounds of shear modulus of Hashin [Hashin, 1962] and Jayasundere's model [Jayasundere et. al., 1994]. Experimental data are taken from [Zou et. al., 1996] of Zou et al..

To conclude, our scheme and Wong et al. scheme [Wong et. al., 2001] provide two different approaches for the predictions of the effective piezoelectric strain coefficients. While Wong et. al.'s scheme can only provide the lower and the upper bound values, our scheme can give reasonable predictions for the whole range of the volume fractions of the inclusions.

2.4 Explicit formulae of the effective piezoelectric coefficients

2.4.1 Theory

In the following, we shall further extend the idea used by Poon and Shin for treating the piezoelectric problems of a 0-3 composite.

Consider a composite with volume fraction ϕ of the piezoelectric inclusions subjected to external stresses T_1 , T_2 and T_3 , and an electric field E_3 applied in the Z direction.

For electrical properties, because the applied electric field can induce strain inside the constituents, we consider that both constituents are piezoelectrically transversely isotropic. In this case, the piezoelectric behaviour of the constituents is described by the following relation [ANSI/IEEE Std., 176-1987],

$$\begin{pmatrix} T_{11p} \\ T_{22p} \\ T_{33p} \\ D_{3p} \end{pmatrix} = \begin{pmatrix} C_{11p} & C_{12p} & C_{12p} & -e_{31p} \\ C_{12p} & C_{11p} & C_{12p} & -e_{31p} \\ C_{12p} & C_{12p} & C_{11p} & -e_{33p} \\ e_{31p} & e_{31p} & e_{33p} & \varepsilon_p \end{pmatrix} \begin{pmatrix} S_{11p} \\ S_{22p} \\ S_{33p} \\ E_{3p} \end{pmatrix}$$
(2.41)

Based on Poon and Shin idea and using equation (2.41), equation (2.5) can be modified to the following

$$\Delta D = -2\varepsilon_m \Delta E + \phi(\varepsilon_i - \varepsilon_m) \langle E_{3i} \rangle + \phi(e_{31i} - e_{31m}) \langle S_{11i} \rangle$$
$$+ \phi(e_{31i} - e_{31m}) \langle S_{22i} \rangle + \phi(e_{33i} - e_{33m}) \langle S_{33i} \rangle \qquad (2.42)$$

where e_{31p} and e_{33p} are the transverse and longitudinal piezoelectric stress coefficients respectively.

For mechanical properties, when external stresses are applied to the piezocomposite, electric fields are induced inside the constituents. Using the Poon and Shin idea and the constitute piezoelectric relation, we obtain new δT_{m1} , δT_{m2} and δT_{m3} expressions, as follows

$$\delta T_{m1} = \frac{1}{V} \int_{V} [(T_{11} - (C_{11m}S_{11} + C_{12m}S_{22} + C_{12m}S_{33} - e_{31m}E_{3m})] dV$$
$$= \phi (\Delta C_{11} \langle S_{11i} \rangle + \Delta C_{12} \langle S_{22i} \rangle + \Delta C_{12} \langle S_{33i} \rangle - \Delta e_{31} \langle E_{3i} \rangle) \qquad (2.43)$$

$$\delta T_{m2} = \frac{1}{V} \int_{V} [(T_{22} - (C_{12m}S_{11} + C_{11m}S_{22} + C_{12m}S_{33} - e_{31m}E_{3m})]dV$$
$$= \phi (\Delta C_{12} \langle S_{11i} \rangle + \Delta C_{11} \langle S_{22i} \rangle + \Delta C_{12} \langle S_{33i} \rangle - \Delta e_{31} \langle E_{3i} \rangle) \qquad (2.44)$$

$$\delta T_{m2} \equiv \frac{1}{V} \int_{V} [(T_{33} - (C_{12m}S_{11} + C_{12m}S_{22} + C_{11m}S_{33} - e_{33m}E_{3m})] dV$$
$$\equiv \phi (\Delta C_{12} \langle S_{11i} \rangle + \Delta C_{12} \langle S_{22i} \rangle + \Delta C_{11} \langle S_{33i} \rangle - \Delta e_{33} \langle E_{3i} \rangle) \qquad (2.45)$$

where

$$\Delta e_{31} \equiv e_{31i} - e_{31m}, \quad \Delta e_{33} \equiv e_{33i} - e_{33m}$$
$$\Delta C_{11} \equiv C_{11i} - C_{11m}, \quad \Delta C_{12} \equiv C_{12i} - C_{12m}$$
$$C_{11p} = k_p + \frac{4}{3}\mu_p, \quad C_{12p} = k_p - \frac{2}{3}\mu_p$$

Equation (2.19) is modified to the following

$$\begin{pmatrix} \Delta T_1 \\ \Delta T_2 \\ \Delta T_3 \end{pmatrix} = \begin{pmatrix} A & B & B \\ B & A & B \\ B & B & A \end{pmatrix} \begin{pmatrix} \Delta S_1 \\ \Delta S_2 \\ \Delta S_3 \end{pmatrix} + \begin{pmatrix} \delta T_{m1} \\ \delta T_{m2} \\ \delta T_{m3} \end{pmatrix}$$
(2.46)

Combining equations (2.42) and (2.46), we have

$$\begin{pmatrix} \Delta T_{1} \\ \Delta T_{2} \\ \Delta T_{3} \\ \Delta D_{3} \end{pmatrix} = \begin{pmatrix} A & B & B & 0 \\ B & A & B & 0 \\ B & B & A & 0 \\ 0 & 0 & 0 & -2\varepsilon_{m} \end{pmatrix} \begin{pmatrix} \Delta S_{1} \\ \Delta S_{2} \\ \Delta S_{3} \\ \Delta E_{3} \end{pmatrix} + \phi \begin{pmatrix} \Delta C_{11} & \Delta C_{12} & \Delta C_{12} & -\Delta e_{31} \\ \Delta C_{12} & \Delta C_{11} & \Delta C_{12} & -\Delta e_{31} \\ \Delta C_{12} & \Delta C_{12} & \Delta C_{11} & -\Delta e_{33} \\ \Delta e_{31} & \Delta e_{31} & \Delta e_{33} & \varepsilon_{i} - \varepsilon_{m} \end{pmatrix} \begin{pmatrix} \langle S_{11i} \rangle \\ \langle S_{22i} \rangle \\ \langle S_{33i} \rangle \\ \langle E_{3i} \rangle \end{pmatrix}$$

$$(2.47)$$

From the definition of the volumetric averages and using equation (2.41), we

have

$$\begin{pmatrix} \langle S_{11} \rangle \\ \langle S_{22} \rangle \\ \langle S_{33} \rangle \\ \langle E_3 \rangle \end{pmatrix} = \phi \begin{pmatrix} \langle S_{11i} \rangle \\ \langle S_{22i} \rangle \\ \langle S_{33i} \rangle \\ \langle E_{i3} \rangle \end{pmatrix} + (1 - \phi) \begin{pmatrix} \langle S_{11m} \rangle \\ \langle S_{22m} \rangle \\ \langle S_{33m} \rangle \\ \langle E_{m3} \rangle \end{pmatrix}$$
(2.48)

$$\begin{pmatrix} \langle T_{1} \rangle \\ \langle T_{2} \rangle \\ \langle T_{3} \rangle \\ \langle D_{3} \rangle \end{pmatrix} = \phi \begin{pmatrix} C_{11i} & C_{12i} & C_{12i} & -e_{31i} \\ C_{12i} & C_{12i} & C_{11i} & -e_{33i} \\ e_{31i} & e_{31i} & e_{33i} & \varepsilon_{i} \end{pmatrix} \begin{pmatrix} \langle S_{11i} \rangle \\ \langle S_{22i} \rangle \\ \langle S_{33i} \rangle \\ \langle E_{3i} \rangle \end{pmatrix} + (1-\phi) \begin{pmatrix} C_{11m} & C_{12m} & C_{12m} & -e_{31m} \\ C_{12m} & C_{11m} & C_{12m} & -e_{31m} \\ C_{12m} & C_{11m} & -e_{33m} \\ e_{31m} & e_{31m} & e_{33m} & \varepsilon_{m} \end{pmatrix} \begin{pmatrix} \langle S_{11m} \rangle \\ \langle S_{22m} \rangle \\ \langle S_{33m} \rangle \\ \langle E_{3m} \rangle \end{pmatrix}$$

$$(2.49)$$

Using equation (2.47) and the constitutive piezoelectric relations, we get

$$C_{i}\langle S_{i}\rangle - C_{m}\langle S_{m}\rangle = L(\langle S_{i}\rangle - \langle S_{m}\rangle) + \phi M\langle S_{i}\rangle$$
(2.50)

where

$$C_{p} \equiv \begin{pmatrix} C_{11p} & C_{12p} & C_{12p} & -e_{31p} \\ C_{12p} & C_{11p} & C_{12p} & -e_{31p} \\ C_{12p} & C_{12p} & C_{11p} & -e_{33z} \\ e_{31p} & e_{31p} & e_{33p} & \varepsilon_{p} \end{pmatrix}, \quad \langle S_{p} \rangle \equiv \begin{pmatrix} \langle S_{11p} \rangle \\ \langle S_{22p} \rangle \\ \langle S_{33p} \rangle \\ \langle E_{3p} \rangle \end{pmatrix}$$
$$L \equiv \begin{pmatrix} A & B & B & 0 \\ B & A & B & 0 \\ B & B & A & 0 \\ 0 & 0 & 0 & -2\varepsilon_{m} \end{pmatrix}, \quad M \equiv \begin{pmatrix} \Delta C_{11} & \Delta C_{12} & \Delta C_{12} & -\Delta e_{31} \\ \Delta C_{12} & \Delta C_{11} & \Delta C_{12} & -\Delta e_{31} \\ \Delta C_{12} & \Delta C_{11} & -\Delta e_{33} \\ \Delta C_{12} & \Delta C_{11} & -\Delta e_{33} \\ \Delta e_{31} & \Delta e_{31} & \Delta e_{33} & \varepsilon_{i} - \varepsilon_{m} \end{pmatrix}$$

After rearrangement, we obtain

$$[C_i - (L + \phi M)] \langle S_i \rangle = (C_m - L) \langle S_m \rangle$$
(2.51)

Multiplying $[C_i - (L + \phi M)]$ on both sides of equation (2.48), we get

$$[C_{i} - (L + \phi M)] \langle S \rangle = \phi [C_{i} - (L + \phi M)] \langle S_{i} \rangle + (1 - \phi) [C_{i} - (L + \phi M)] \langle S_{m} \rangle$$

$$(2.52)$$

$$(\langle S_{i} \rangle)$$

where
$$\langle S \rangle \equiv \begin{vmatrix} \langle S_1 \rangle \\ \langle S_2 \rangle \\ \langle S_3 \rangle \\ \langle E_3 \rangle \end{vmatrix}$$

Using equation (2.51), equation (2.52) becomes

$$[C_{i} - (L + \phi M)]\langle S \rangle = \phi(C_{m} - L)\langle S_{m} \rangle + (1 - \phi)[C_{i} - (L + \phi M)]\langle S_{m} \rangle$$

$$(2.53)$$

After simplifying, we have

$$\langle S_m \rangle = \{ \phi(C_m - L) + (1 - \phi)[C_i - (L + \phi M)] \}^{-1} [C_i - (L + \phi M)] \langle S \rangle$$

(2.54)

Multiplying $(C_m - L)$ on both sides of equation (2.48), we get

$$(C_m - L)\langle S \rangle = \phi(C_m - L)\langle S_i \rangle + (1 - \phi)(C_m - L)\langle S_m \rangle$$
(2.55)

Using equation (2.51), equation (2.55) becomes

$$(C_m - L)\langle S \rangle = \phi(C_m - L)\langle S_i \rangle + (1 - \phi)[C_i - (L + \phi M)]\langle S_i \rangle \qquad (2.56)$$

After simplifying, we have

$$\langle S_i \rangle = \{ \phi(C_m - L) + (1 - \phi) [C_i - (L + \phi M)] \}^{-1} (C_m - L) \langle S \rangle$$

(2.57)

Substituting equations (2.54) and (2.57) into equation (2.49), we get

$$\langle T \rangle = \phi C_i \langle S_i \rangle + (1 - \phi) C_m \langle S_m \rangle$$

$$= \{ \phi C_i \{ \phi (C_m - L) + (1 - \phi) [C_i - (L + \phi M)] \}^{-1} (C_m - L)$$

$$+ (1 - \phi) C_m \{ \phi (C_m - L) + (1 - \phi) [C_i - (L + \phi M)] \}^{-1} [C_i - (L + \phi M)] \} \langle S \rangle$$

$$(2.58)$$

where
$$\langle T \rangle \equiv \begin{pmatrix} \langle T_1 \rangle \\ \langle T_2 \rangle \\ \langle T_3 \rangle \\ \langle D_3 \rangle \end{pmatrix}$$

After simplifying, we get, in matrix form

$$\begin{pmatrix} \langle T_1 \rangle \\ \langle T_2 \rangle \\ \langle T_3 \rangle \\ \langle D_3 \rangle \end{pmatrix} = \begin{pmatrix} C_{11} & C_{12} & C_{13} & -e_{31} \\ C_{12} & C_{11} & C_{13} & -e_{31} \\ C_{13} & C_{13} & C_{33} & -e_{33} \\ e_{31} & e_{31} & e_{33} & \varepsilon \end{pmatrix} \begin{pmatrix} \langle S_1 \rangle \\ \langle S_2 \rangle \\ \langle S_3 \rangle \\ \langle E_3 \rangle \end{pmatrix}$$
(2.59)

where the effective piezoelectric and mechanical coefficients are given by

$$e_{31} = \phi [I_{41i}(C_{11m} - A + C_{12m} - B) + I_{43i}(C_{12m} - B) + I_{44i}e_{31m}]$$

+ $(1 - \phi) [I_{41m}(C_{11i} - \phi \Delta C_{11} - A + C_{12i} - \phi \Delta C_{12} - B)$
+ $I_{43m}(C_{12i} - \phi \Delta C_{12} - B) + I_{44m}(e_{31i} - \phi \Delta e_{31})]$ (2.60)

$$e_{33} = \phi [2I_{41i}(C_{12m} - B) + I_{43i}(C_{11m} - A) + I_{44i}e_{33m}] + (1 - \phi) [2I_{41m}(C_{12i} - \phi\Delta C_{12} - B) + I_{43m}(C_{11i} - \phi\Delta C_{11} - A) + I_{44m}(e_{33i} - \phi\Delta e_{33})]$$

$$(2.61)$$

$$C_{11} = \phi [I_{11i}(C_{11m} - A) + (I_{12i} + I_{13i})(C_{12m} - B) + I_{14i}e_{31m}]$$

+ $(1 - \phi)[I_{11m}(C_{11i} - \phi\Delta C_{11} - A) + (I_{12m} + I_{13m})(C_{12i} - \phi\Delta C_{12} - B)$
+ $I_{14m}(e_{31i} - \phi\Delta e_{31})]$ (2.62)

$$C_{12} = \phi [I_{12i}(C_{11m} - A) + (I_{11i} + I_{13i})(C_{12m} - B) + I_{14i}e_{31m}]$$

+ $(1 - \phi)[I_{12m}(C_{11i} - \phi\Delta C_{11} - A) + (I_{11m} + I_{13m})(C_{12i} - \phi\Delta C_{12} - B)$
+ $I_{14m}(e_{31i} - \phi\Delta e_{31})]$ (2.63)

$$C_{13} = \phi [I_{13i}(C_{11m} - A) + (I_{11i} + I_{12i})(C_{12m} - B) + I_{14i}e_{33m}]$$

+ $(1 - \phi)[I_{13m}(C_{11i} - \phi \Delta C_{11} - A) + (I_{11m} + I_{12m})(C_{12i} - \phi \Delta C_{12} - B)$
+ $I_{14m}(e_{33i} - \phi \Delta e_{33})]$ (2.64)

$$C_{33} = \phi [I_{33i}(C_{11m} - A) + (I_{31i} + I_{32i})(C_{12m} - B) + I_{34i}e_{33m}]$$

+ $(1 - \phi)[I_{33m}(C_{11i} - \phi \Delta C_{11} - A) + (I_{31m} + I_{32m})(C_{12i} - \phi \Delta C_{12} - B)$
+ $I_{34m}(e_{33i} - \phi \Delta e_{33})]$ (2.65)

The symbols I_{klp} in equations (2.60) to (2.65) are defined below, in which the

subscripts k, l = 1, 2, 3 refer to the X, Y and Z directions.

$$\begin{pmatrix} I_{11p} & I_{12p} & I_{13p} & I_{14p} \\ I_{12p} & I_{11p} & I_{13p} & I_{14p} \\ I_{31p} & I_{31p} & I_{33p} & I_{34p} \\ I_{41p} & I_{41p} & I_{43p} & I_{44p} \end{pmatrix} = \begin{pmatrix} C_{11p} & C_{12p} & C_{12p} & -e_{31p} \\ C_{12p} & C_{11p} & C_{12p} & -e_{31p} \\ C_{12p} & C_{12p} & C_{11p} & -e_{33p} \\ e_{31p} & e_{31p} & e_{33p} & \varepsilon_p \end{pmatrix} \begin{pmatrix} I_{11} & I_{12} & I_{13} & -I_{14} \\ I_{12} & I_{11} & I_{13} & -I_{14} \\ I_{13} & I_{13} & I_{33} & -I_{34} \\ I_{14} & I_{14} & I_{34} & I_{44} \end{pmatrix}$$

where

$$\begin{pmatrix} I_{11} & I_{12} & I_{13} & -I_{14} \\ I_{12} & I_{11} & I_{13} & -I_{14} \\ I_{13} & I_{13} & I_{33} & -I_{34} \\ I_{14} & I_{14} & I_{34} & I_{44} \end{pmatrix} = \begin{cases} \phi \begin{pmatrix} C_{11m} - A & C_{12m} - B & C_{12m} - B & -e_{31m} \\ C_{12m} - B & C_{11m} - A & C_{12m} - B & -e_{31m} \\ C_{12m} - B & C_{12m} - B & C_{11m} - A & -e_{33m} \\ e_{31m} & e_{31m} & e_{33m} & 3\mathcal{E}_m \end{pmatrix} \\ + (1-\phi) \begin{pmatrix} C_{11i} - \phi \Delta C_{11} - A & C_{12i} - \phi \Delta C_{12} - B & C_{12i} - \phi \Delta C_{12} - B & -e_{31i} + \phi \Delta e_{31} \\ C_{12i} - \phi \Delta C_{12} - B & C_{11i} - \phi \Delta C_{11} - A & C_{12i} - \phi \Delta C_{12} - B & -e_{31i} + \phi \Delta e_{31} \\ C_{12i} - \phi \Delta C_{12} - B & C_{11i} - \phi \Delta C_{11} - A & C_{12i} - \phi \Delta C_{12} - B & -e_{31i} + \phi \Delta e_{31} \\ C_{12i} - \phi \Delta C_{12} - B & C_{11i} - \phi \Delta C_{11} - A & C_{12i} - \phi \Delta C_{12} - B & -e_{31i} + \phi \Delta e_{31} \\ C_{12i} - \phi \Delta C_{12} - B & C_{12i} - \phi \Delta C_{12} - B & C_{11i} - \phi \Delta C_{11} - A & -e_{33i} + \phi \Delta e_{33} \\ e_{31i} - \phi \Delta e_{31} & e_{31i} - \phi \Delta e_{31} & e_{33i} - \phi \Delta e_{33} & \mathcal{E}_i + 2\mathcal{E}_m - \phi(\mathcal{E}_m - \mathcal{E}_i) \end{pmatrix} \right\}^{-1}$$

Combing the two effective piezoelectric stress coefficients and the four stiffness constants, two effective piezoelectric strain coefficients (d_{31} and d_{33})

are obtained. For a transversely isotropic material, the relation are given by

$$d_{31} = \frac{\frac{e_{31}}{C_{13}} - \frac{e_{33}}{C_{33}}}{\frac{C_{11} + C_{12}}{C_{13}} - \frac{2C_{13}}{C_{33}}}$$
(2.66)
$$d_{33} = \frac{\frac{e_{31}}{C_{11} + C_{12}} - \frac{e_{33}}{2C_{13}}}{\frac{C_{13}}{C_{11} + C_{12}} - \frac{C_{33}}{2C_{13}}}$$
(2.67)

In the following section, our model is compared with some experimental results. Wong et al.'s scheme [Wong et. al., 2001] is also included in the comparisons. They have assumed that the whole composite is elastically isotropic. If we also take this assumption (i.e. $C_{33} = C_{11}$ and $C_{13} = C_{12}$), then the two effective piezoelectric strain coefficients (d_{31} and d_{33}) become:

$$d_{31} = \frac{\frac{e_{31}}{C_{12}} - \frac{e_{33}}{C_{11}}}{\frac{C_{11} + C_{12}}{C_{12}} - \frac{2C_{12}}{C_{11}}}$$
(2.68)
$$d_{33} = \frac{\frac{e_{31}}{C_{11} + C_{12}} - \frac{e_{33}}{2C_{12}}}{\frac{C_{12}}{C_{11} + C_{12}} - \frac{C_{11}}{2C_{12}}}$$
(2.69)

2.4.2 Comparisons with experimental data and discussions

In this subsection, predictions made by Wong et al. [Wong et. al., 2001] and our model (with and without assuming elastic isotropy) are compared with the experimental data of Furukawa [Furukawa, 1989], Chan et al. [Chan et. al., 1995] and Zeng et al. [Zeng et. al., 2002] for the d_{31} of PZT/PVDF composites (with only the ceramic phase polarized), the d_{33} of PZT/P(VDF-TrFE) composites (with both phases polarized in the same direction) and d_{31} , d_{33} of PZT/P(VDF-TrFE) composites (with the two phases polarized in opposite directions). The parameters of the constituents

adopted in the calculations are listed in tables 2.1 to 2.3. Elastic moduli Y_p and Poisson's ratios v_p in tables 2.1 and 2.2 can be transformed to obtain the stiffness constants C_{11p} and C_{12p} via the relations [Saada, 1993]

$$C_{11p} = \frac{Y_p}{3(1 - 2v_p)} + \frac{2}{3} \frac{Y_p}{(1 + v_p)}$$
(2.70)

$$C_{12p} = \frac{Y_p}{3(1 - 2v_p)} - \frac{1}{3} \frac{Y_p}{(1 + v_p)}$$
(2.71)

The constituents' parameters of shear moduli μ_p and Poisson's ratios ν_p in table 2.3 can be transformed to obtain the stiffness constants C_{11p} and C_{12p} via the following relations [Saada, 1993]

$$C_{11p} = \frac{2(1+\nu_p)\mu_p}{3(1-2\nu_p)} + \frac{4}{3}\mu_p$$
(2.72)

$$C_{12p} = \frac{2(1+\nu_p)\mu_p}{3(1-2\nu_p)} - \frac{2}{3}\mu_p$$
(2.73)

Elastic Poisson's $-d_{31}$ d_{33} $\mathcal{E}/\mathcal{E}_0$ Modulus Ratio (pC/N)(pC/N)(GPa) PZT 1159 16.8 0.35 127.9 314.4 P(VDF-10.7 2.32 0.39 -15.3 -38.4 TrFE)

Table 2.1. Properties of the constituents for PZT/P(VDF-TrFE) 0-3 composite adopted in our calculations in Fig. 2.7. Both phases are polarized in the same direction.[Wong et. al., 2001].

Table 2.2. Properties of the constituents for PZT/PVDF 0-3 composite adopted in our calculations in Fig. 2.8. Only the inclusion phase is polarized.[Wong et. al., 2005]

	$\varepsilon/\varepsilon_0$	Elastic Modulus (GPa)	Poisson's Ratio	- d ₃₁ (pC/N)	<i>d</i> ₃₃ (pC/N)
PZT	1900	36	0.3	180	450
PVDF	14	1.3	0.4	0	0

Table 2.3 Properties of the constituents for PZT/P(VDF-TrFE) 0-3 compositeadopted in our calculations in Fig. 2.9. [Zeng et. al., 2002]

	$\varepsilon/\varepsilon_0$	Shear Modulus (GPa)	Poisson's Ratio	- d ₃₁ (pC/N)	d ₃₃ (pC/N)
PZT	1700	27	0.3	175	400
P(VDF- TrFE)	9.9	0.8	0.4	-6.2	-22.1

Figure 2.7 shows the d_{33} comparison results of the theoretical predictions and the experimental data of Chan et al [Chan et. al., 1995]. Note that because the piezoelectric coefficients of PZT and P(VDF-TrFE) have opposite signs, the piezoelectric activity of the composite vanishes at $\phi \approx 0.45$ when both phases are polarized in the same direction.

Obviously, both our model and Wong et al's scheme give similar reasonable performance.



Figure 2.7 Predictions of effective piezoelectric coefficient d₃₃ by our model (equations (2.67) and (2.69)) and Wong et al.'s scheme [Wong et. al., 2001] with the experimental data of H.L.W Chan et al. [Chan et. al., 1995] of PZT/P(VDF-TrFE) composite, with inclusion and matrix polarized in the same direction.

Figure 2.8 shows the d_{31} comparison. In this composite material, the effective piezoelectric activity is contributed by the inclusion phase only. It can be seen that, at low volume fractions, both our model and Wong et al.'s scheme give similar performance. However, at high volume fractions, some of the experimental data are not even within the bounds calculated by Wong et al's scheme but our model still fits well to the experimental data.



Figure 2.8 Predictions of effective piezoelectric coefficient d₃₁ by our model (equations (2.66) and (2.68)) and Wong et al.'s scheme [Wong et. al., 2001] with the experimental data of Furukawa [Furukawa, 1989] of PZT/PVDF composites. Only the inclusion phase is polarized.

Zeng et al [Zeng et. al., 2002] reported experimental values of the piezoelectric coefficients d_{31} and d_{33} of a PZT/P(VDF-TrFE) composite, with the two phases polarized in opposite directions. The comparisons of the experimental data with some theoretical predictions are shown in Figure 2.9. When compared with Wong et al.'s scheme, our model obviously gives more reasonable predictions to the experimental data. As a research, the abrupt decrease of the piezoelectric coefficient value at $\phi \approx 0.5$, according to Zeng et. al., may be due to the redistribution of space charges at the interface between the inclusion and the matrix. These changes could lead to some degree of depolarization and hence decrease the piezoelectric activity.

Since both our model and Wong et al's scheme have assumed that the constituents are fully polarized, this may be the reason why both models show deviations to the experimental values, at high volume fractions of the inclusions.



Figure 2.9 Predictions of effective piezoelectric coefficients d₃₃ (equations (2.67) and (2.69)) and d₃₁ (equations (2.66) and (2.68)) by our model and Wong et al.'s scheme [Wong et. al., 2001] with the experimental data of Zeng et al. [Zeng et. al. 2002] of PZT/P(VDF-TrFE) composite with inclusion and matrix polarized in opposite directions.

From the above comparisons, it also shows that the two different assumptions (isotropic or transversely isotropic) concerning the mechanical properties of the two phases give similar performance. This means that the effects of polarization on the elasticity of the materials are very small.
Chapter 3 Effective piezoelectric coefficients of 1-3 composites based on an effective medium theory

3.1 Introduction

1-3 piezoelectric composites usually consist of piezoelectric rods or fibers embedded in a polymer matrix. They have been widely used in ultrasonic transducers in underwater and other ultrasonic medical applications [Smith, 1989]. The polymer phase can be either a passive (e.g. epoxy) or an active (e.g. PVDF-TrFE) matrix medium [Taunaumang et. al., 1994]. For the former, the polymer phase is responsible only for stress transfer to the inclusion phase. For the latter, the effective piezoelectric properties of the composite are contributed by both the inclusion phase and the polymer phase. Due to the opposite signs of their piezoelectric activities, it is possible to pole the two phases in opposite directions to increase the effective piezoelectric coefficients d_{31} and d_{31} of the composite material [Taunaumang et. al., 1994]. In this work, we consider the former case only, since it is the mostly used case in actual applications.

In this chapter, we consider the piezoelectric problems of 1-3 piezoelectric rod composite having small aspect ratios. Its effective piezoelectric coefficients are determined based on an effective medium theory (EMT). This theory considers the volume fraction of the inclusions (ϕ) and the matrix properties as independent variables, and then expressions of the effective properties for the high volume fraction cases can be obtained from the dilute limit results. The EMT formulation employed in this project was first developed by Shin et. al. [Shin et. al., 1989] and has been used in treating the dielectric [Shin et. al., 1989], piezoelectric [Wong et. al., 2003] and pyroelectric [Chew et. al., 2003] problems of binary 0-3 composites. EMT equations can be either solved analytically or numerically. The comparisons of the predictions made by analytical EMT and numerical EMT and the published data are presented in the results and discussions section. In the following, we use the subscripts i and m to represent the inclusion phase and matrix phase respectively and use the symbol p to refer either ior m. The subscripts 1, 2 and 3 denote the X, Y and Z directions respectively.

3.2.1 An effective medium theory (EMT)

The EMT formulation used in this project was first developed by Shin et. al. [Shin et. al., 1989]. Suppose we have a binary composite with an effective physical property P. This composite property P should be a function of the matrix property P_m , the inclusion property P_i and the volume fraction of the inclusions ϕ . That is

$$P = f(P_m, P_i, \phi) \tag{3.1}$$

Suppose we start with a pure matrix with the physical property P_m . Now, we add some inclusions with the physical property P_i into it. Assuming that the volume fraction of the inclusions of this resulting composite at this stage is ϕ_1 . The effective property P_1 should be given by

$$P_1 = f(P_m, P_i, \phi_1)$$
(3.2)

Now, we take this composite as a new matrix and we add inclusions into this new matrix such that the volume fraction with respect to this new matrix is ϕ_2 . The effective property P_2 of the resulting composite is then

$$P_{2} = f(P_{1}, P_{i}, \phi_{2})$$

= $f(f(P_{m}, P_{i}, \phi_{1}), P_{i}, \phi_{2})$ (3.3)

If the actual volume fraction of this composite is ϕ , we must have

$$f(P_m, P_i, \phi) = f(f(P_m, P_i, \phi_1), P_i, \phi_2)$$
(3.4)

and

$$\phi = \phi_2 + (1 - \phi_2)\phi_1 \tag{3.5}$$

Differentiating equation (3.4) with respect to ϕ_1 , then setting $\phi_1 = 0$, we get

$$(1-\phi)\frac{\partial f(P_m, P_i, \phi)}{\partial \phi} = \frac{\partial f(P_m, P_i, \phi)}{\partial P_m} \left[\frac{\partial f(P_m, P_i, \phi)}{\partial \phi}\right]_{\phi=0}$$
(3.6)

Equation (3.6) is the first order partial differential equation which involves one matrix variable only. In the following subsection, we use the same idea, but take into account more matrix variables, in order to find the effective piezoelectric coefficients of 1-3 composites.

3.2.2 Effective piezoelectric stress coefficients of a 1-3 composite based on analytical EMT approach

As described in the previous section, EMT is an approach to obtain expressions of the effective properties at high volume fraction cases based on the dilute limit results. Therefore, before applying this approach, we have to consider the piezoelectric problem of a 1-3 composite with low volume fraction of inclusions. Suppose we have a composite with a single piezoelectric fiber embedded in a piezoelectric matrix and the fiber (and hence the composite) is poled along its axial direction (Z directions). We assume that both phases are transversely isotropic materials. Under proper boundary conditions, expressions of the effective piezoelectric stress coefficients (e_{33} and e_{31}) and the stiffness constants (C_{11} , C_{12} and C_{13}) can be found, as follows (see Appendix C)

$$e_{31} = \frac{\phi e_{31i} \frac{2C_{11m}}{C} + (1-\phi)e_{31m}}{\phi \frac{2C_{11m}}{C} + 1 - \phi}$$
(3.7)

$$e_{33} = \phi e_{33i} + (1 - \phi)e_{33m} + 2\frac{C_{13m} - C_{13i}}{C}\frac{\phi(1 - \phi)}{\phi\frac{2C_{11m}}{C} + 1 - \phi}(e_{31i} - e_{31m})$$
(3.8)

$$C_{11} = \frac{\phi C_{11i} \frac{2C_{11m}}{C} + (1 - \phi)C_{11m}}{\phi \frac{2C_{11m}}{C} + 1 - \phi}$$
(3.9)

$$C_{12} = \frac{\phi C_{12i} \frac{2C_{11m}}{C} + (1 - \phi)C_{12m}}{\phi \frac{2C_{11m}}{C} + 1 - \phi}$$
(3.10)

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$$C_{13} = \frac{\phi C_{13i} \frac{2C_{11m}}{C} + (1 - \phi)C_{13m}}{\phi \frac{2C_{11m}}{C} + 1 - \phi}$$
(3.11)

where $C \equiv C_{11i} + C_{12i} + C_{11m} - C_{12m}$

and $C_{\alpha\beta p}$, e_{31p} and e_{33p} are stiffness constants, transverse piezoelectric stress coefficients and longitudinal piezoelectric stress coefficients of the constituents respectively.

Now we apply the EMT approach to find the effective piezoelectric stress coefficients for non-dilute cases. The expressions (3.7), (3.8) show that e_{33} and e_{31} involve five and three matrix parameters respectively. By taking analogy to equation (3.6), the first order partial differential equations that e_{33} and e_{31} should satisfy

$$(1-\phi)\frac{\partial e_{33}}{\partial \phi} = \frac{\partial e_{33}}{\partial e_{33m}} \left[\frac{\partial e_{33}}{\partial \phi} \right]_{\phi=0} + \frac{\partial e_{33}}{\partial e_{31m}} \left[\frac{\partial e_{31}}{\partial \phi} \right]_{\phi=0} + \frac{\partial e_{33}}{\partial C_{11m}} \left[\frac{\partial C_{11}}{\partial \phi} \right]_{\phi=0} + \frac{\partial e_{33}}{\partial C_{12m}} \left[\frac{\partial C_{12}}{\partial \phi} \right]_{\phi=0} + \frac{\partial e_{33}}{\partial C_{12m}} \left[\frac{\partial C_{12}}{\partial \phi} \right]_{\phi=0}$$

(3.12)

$$(1-\phi)\frac{\partial e_{31}}{\partial \phi} = \frac{\partial e_{31}}{\partial e_{31m}} \left[\frac{\partial e_{31}}{\partial \phi}\right]_{\phi=0} + \frac{\partial e_{31}}{\partial C_{11m}} \left[\frac{\partial C_{11}}{\partial \phi}\right]_{\phi=0} + \frac{\partial e_{31}}{\partial C_{12m}} \left[\frac{\partial C_{12}}{\partial \phi}\right]_{\phi=0}$$
(3.13)

The characteristic equations of (3.12) and (3.13) are (3.14) and (3.15), respectively.

$$\frac{-d\phi}{1-\phi} = \frac{de_{33m}}{\left[\partial e_{33} / \partial \phi\right]_{\phi=0}} = \frac{de_{31m}}{\left[\partial e_{31} / \partial \phi\right]_{\phi=0}} = \frac{dC_{11m}}{\left[\partial C_{11} / \partial \phi\right]_{\phi=0}}$$
$$= \frac{dC_{12m}}{\left[\partial C_{12} / \partial \phi\right]_{\phi=0}} = \frac{dC_{13m}}{\left[\partial C_{13} / \partial \phi\right]_{\phi=0}}$$
(3.14)

$$\frac{-d\phi}{1-\phi} = \frac{de_{_{31m}}}{\left[\partial e_{_{31}} / \partial \phi\right]_{\phi=0}} = \frac{dC_{_{11m}}}{\left[\partial C_{_{11}} / \partial \phi\right]_{\phi=0}} = \frac{dC_{_{12m}}}{\left[\partial C_{_{12}} / \partial \phi\right]_{\phi=0}}$$
(3.15)

The derivatives of the effective stiffness constants and the effective piezoelectric stress coefficients of the composite with respect to ϕ at the dilute limit can be obtained from expressions (3.7) to (3.11), as follows

$$\begin{split} \left[\frac{\partial C_{11}}{\partial \phi}\right]_{\phi=0} &= \frac{2C_{11m}}{C}(C_{11i} - C_{11m})\\ \left[\frac{\partial C_{12}}{\partial \phi}\right]_{\phi=0} &= \frac{2C_{11m}}{C}(C_{12i} - C_{12m})\\ \left[\frac{\partial C_{13}}{\partial \phi}\right]_{\phi=0} &= \frac{2C_{11m}}{C}(C_{13i} - C_{13m})\\ \left[\frac{\partial e_{31}}{\partial \phi}\right]_{\phi=0} &= \frac{2C_{11m}}{C}(e_{31i} - e_{31m})\\ \left[\frac{\partial e_{33}}{\partial \phi}\right]_{\phi=0} &= e_{33i} - e_{33m} - \frac{2(C_{13i} - C_{13m})}{C}(e_{31i} - e_{31m}) \end{split}$$

From equation (3.15), we can write

$$\frac{de_{31m}}{\left[\partial e_{31} / \partial \phi\right]_{\phi=0}} = \frac{dC_{11m} + dC_{12m}}{\left[\partial C_{11} / \partial \phi\right]_{\phi=0} + \left[\partial C_{12} / \partial \phi\right]_{\phi=0}}$$
(3.16)

After integration, we get

$$e_{31} = e_{31i} - (e_{31i} - e_{31m}) \frac{C_{11i} - C_{11} + C_{12i} - C_{12}}{C_{11i} - C_{11m} + C_{12i} - C_{12m}}$$
(3.17)

The equalities in equation (3.15) linking e_{31} , C_{11} and C_{12} can be integrated to yield a first integral

$$\gamma = \frac{e_{31i} - e_{31m}}{C_{11i} - C_{11m} + C_{12i} - C_{12m}}$$
(3.18)

Another first integral can be obtained by considering the equality in equation (3.14) that links e_{31} and C_{13}

$$\omega = \frac{C_{13i} - C_{13m}}{e_{31i} - e_{31m}}$$
(3.19)

With the use of the equalities in equation (3.14) and the first integrals γ and ω , we get

$$\frac{-d\phi}{1-\phi} = \frac{de_{33m} - 2\gamma\omega de_{31m}}{\left[\partial e_{33} / \partial \phi\right]_{\phi=0} - 2\gamma\omega \left[\partial e_{31} / \partial \phi\right]_{\phi=0}}$$
(3.20)

Equation (3.20) can be integrated to give

$$e_{33} = e_{33i} - 2\gamma\omega(e_{31i} - e_{31}) - (1 - \phi) [(e_{33i} - e_{33m}) - 2\gamma\omega(e_{31i} - e_{31m})]$$
(3.21)

Two analytical EMT expressions of e_{33} and e_{31} (equation (3.17) and (3.21)) are therefore obtained.

3.2.3 Numerical calculation using EMT

Chen [Chen, 1998] outlined the calculation steps of a numerical EMT scheme in solving the elastic problems of 1-3 composite, as follows. For a pure matrix, its volume fraction of inclusion is $\phi_0 = 0$. When a small volume fraction δ of inclusion is added into it, the resulting composite has a volume fraction $\phi_1 = \delta$. The effective modulus of the composite is approximately equal to Effective modulus \approx matrix's modulus + derivates of the effective modulus with respect to volume fraction ϕ evaluated at $\phi = 0$

The above expression is valid only for small volume fraction δ . We take this composite as a new matrix and add the same volume fraction of the inclusions into this new matrix. At this stage, the volume fraction of the composite

 $[\]times \delta$

becomes $\phi_2 = \phi_1 + \delta - \phi_1 \delta$. By repeating this step, the effective modulus at the higher volume fractions can be estimated.

Following these steps, we have estimated the effective piezoelectric properties of 1-3 composites at different volume fractions of the inclusions. Results from both the analytical and numerical EMT schemes are presented in section 3.3.1 and 3.3.2 for the comparisons with some published data.

3.2.4 Effective piezoelectric strain coefficients d_{33} and d_{31}

For transversely isotropic materials, the piezoelectric strain coefficients can be calculated by the following relations [ANSI/IEEE Std., 176-1987].

$$d_{31} = \frac{\frac{e_{31}}{C_{13}} - \frac{e_{33}}{C_{33}}}{\frac{C_{11} + C_{12}}{C_{13}} - \frac{2C_{13}}{C_{33}}}$$
(3.22)
$$d_{33} = \frac{\frac{e_{31}}{C_{11} + C_{12}} - \frac{e_{33}}{2C_{13}}}{\frac{C_{13}}{C_{11} + C_{12}} - \frac{C_{33}}{2C_{13}}}$$
(3.23)

In the analytical EMT calculation, we have adopted Chen model [Chen, 1998]. The transverse bulk modulus (K), transverse shear modulus (G_T) , axial Poisson's ratio (V_A) and axial Young's modulus (E_A) , as given by Chen are

$$\frac{\frac{W}{K_i G_{T_i}^2} (1-\phi)^3 (\frac{3}{G_T} + \frac{2}{K_i} - \frac{1}{G_{T_i}}) - (\frac{1}{G_T} - \frac{1}{G_{T_i}})^3}{(1-\phi)^3 (\frac{1}{G_T} + \frac{1}{K_i})^{\frac{3}{2}}} = \frac{\frac{W}{K_i G_{T_i}^2} (1-\phi)^3 (\frac{3}{G_{T_m}} + \frac{2}{K_i} - \frac{1}{G_{T_i}}) - (\frac{1}{G_{T_m}} - \frac{1}{G_{T_i}})^3}{(\frac{1}{G_{T_m}} + \frac{1}{K_i})^{\frac{3}{2}}}$$

(3.24)

where
$$W \equiv \frac{(K_i - K_m)(G_{Ti} - G_{Tm})^2}{(K_m + G_{Tm})G_{Tm}}$$

$$\frac{(K_i - K)(G_{Ti} - G_T)^2}{(K + G_T)G_T} = \frac{(K_i - K_m)(G_{Ti} - G_{Tm})^2}{(K_m + G_{Tm})G_{Tm}} (1 - \phi)^3$$
(3.25)

$$\frac{K}{(K_i - K)}(v_A - v_{Ai}) = \frac{K_m}{(K_i - K_m)}(v_{Am} - v_{Ai})$$
(3.26)

$$E_{A} = \phi E_{Ai} + (1 - \phi) E_{Am} + \frac{4(\nu_{Am} - \nu_{Ai})^{2}}{(\frac{1}{K_{m}} - \frac{1}{K_{i}})^{2}} (\frac{1 - \phi}{K_{m}} + \frac{\phi}{K_{i}} - \frac{1}{K})$$
(3.27)

Using equations (3.24) - (3.27), the four effective stiffness constants can be determined [Christensen, 1991]

$$C_{11} = K - G_T \tag{3.28}$$

$$C_{12} = K + G_T \tag{3.29}$$

$$C_{13} = 2K\nu_A \tag{3.30}$$

$$C_{33} = E_A + 4\nu_A^2 K \tag{3.31}$$

Using equations (3.17) and (3.21) – (3.31), d_{31} and d_{33} can be determined.

Results and Discussions

3.3.1 Comparisons with experimental data

Predictions based on our formulae are compared with the experimental data of Klicker [Klicker et. al., 1981] for d_{33} of a PZT/Epoxy 1-3 composite. The properties of the constituents adopted in our calculations are listed in Table 3.1.

 Table 3.1 Material parameters of PZT and Epoxy [Klicker et. al., 1981]

 adopted in this calculation

PZT					
C_{11} (GPa)	118.4	C_{12} (GPa)	58.5		
C_{13} (GPa)	59.6	C_{33} (GPa)	102.8		
d_{33} (pC/N)	450	d_{31} (pC/N)	-210		
Epoxy					
C_{11} (GPa)	6.5	C_{12} (GPa)	3.5		

Klicker et. al. [Klicker et. al., 1981] investigated the effects of the inclusion diameter on the effective piezoelectric strain coefficients d_{33} of the composite. They reported experimental data of d_{33} for inclusion diameters $400\mu m$, $600\mu m$ and $840\mu m$. The thickness of these samples was 4mm. The ratio of the diameter of the inclusion to its thickness is called the aspect ratio. In this chapter, we only consider the piezoelectric problem of composite system having small aspect ratios. As the axial and radial displacement functions of a single piezoelectric fiber composite (see Appendix C) are independent of *Z*

coordinate, our EMT models can only be applied for the small aspect ratios composites. For comparison purposes, we choose experimental data of d_{33} corresponding to the smallest inclusion diameter. As shown in fig. 3.1, both analytical and numerical EMT schemes give similar predicted values and they have fairly good agreement with the experimental data.



Figure 3.1 Comparison of theoretical predictions of EMT schemes with the experimental data of Klicker [Klicker et. al., 1981] for d₃₃ of PZT/Epoxy composites

3.3.2 Comparison with simulated data obtained from a finite element method (FEM)

Our formulae are also compared with the simulated data for the effective piezoelectric stress coefficients (e_{31} and e_{33}) of a PZT-5/polymer composite reported by Berger et. al. [Berger et. al., 2005], using numerical finite element method. Figs. 3.2(a) and (b) show the e_{31} and e_{33} comparisons respectively. The material properties of the constituents used in our calculations are shown in Table 3.2.

PZT-5					
C_{11} (GPa)	121.0	C_{12} (GPa)	75.4		
C_{13} (GPa)	75.2	C_{33} (GPa)	111.0		
e_{33} $\left(C/m^2\right)$	15.8	e_{31} $\left(C/m^2\right)$	-5.4		
polymer					
C_{11} (GPa)	3.86	C_{12} (GPa)	2.57		

 Table 3.2 Material parameters of PZT-5 and polymer [Berger et. al., 2005]

 adopted in this calculation

Fig. 3.2(a) shows that equation (3.17) has good performance for dilute suspension cases. At high volume fractions ($\phi > 0.3$), there are tiny discrepancies between the theoretical values and the FEM data. This may be due to the fact that Berger et. al. [Berger et. al., 2005] have calculated the effective properties by using a 1-3 composite having periodic structure. Their composite model consists of spatially periodic representative volume elements (RVEs). Each RVE consists of a single piezoelectric fiber embedded in a polymer matrix. The volume fraction of the 1-3 periodic composite is altered by modifying the diameter of each fiber inside a RVE. This may explain the very small deviations of our values at high volume fractions. However, for e_{33} comparison (shown in Fig. 3.2(b)), equation (3.21) agrees quite well with their simulated values.



(a)



(b)

Figure 3.2 Comparison of theoretical predictions of EMT schemes with the simulated data of Harald Berger et. al. [Berger et. al., 2005] for (a) e_{31} and (b) e_{33} of PZT-5/polymer composites

Chapter 4 *Conclusions*

In this project, I have succeeded in extending Poon and Shin approach [Poon and Shin, 2004] for treating elastic problem and piezoelectric problem of 0-3 composites. In the first part of Chapter 2, I have derived two explicit formulae of the effective stiffness constants. These formulae are then compared with some published experimental data. It demonstrated that the model can give reasonable predictions. There formulae and the explicit formula of the effective dielectric constant reported by Poon and Shin [Poon and Shin, 2004] are then incorporated into Wong et. al.'s scheme [Wong et. al., 2001] for the predictions of d_{31} and d_{33} . Results calculated made by the present scheme, Wong et. al.'s scheme and other theoretical works [Furukawa et. al., 1976, Jayasundere et. al., 1994] are then compared with the experimental data of a PZT/polymer composite [Furukawa, 1989, Zou et. al., 1996]. The comparison showed that the scheme has comparable performance with Wong et. al.'s scheme. In the second part of Chapter 2, assuming that both constituents are dielectrically and elastically isotropic even they are polarized, expressions of two piezoelectric stress coefficients (e_{31} and e_{33}) and four stiffness constants (C_{11} , C_{12} , C_{13} and C_{33}) are derived. Combining with the derived results, two expressions of

effective piezoelectric strain coefficients are obtained. Predictions made by our model and Wong et. al.'s model [Wong et. al., 2001] are compared with the experimental data of a 0-3 PZT/polymer composite system [Furukawa, 1989, Chan et. al., 1995, Zeng et. al., 2002] having three different polarization states: only the inclusion phase is polarized, both phases polarized in the same direction and the two phases polarized in opposite directions. For the first two cases, both our model and Wong et. al.'s model [Wong et. al., 2001] give similar performance. For the last case, when compared with Wong et. al.'s model [Wong et. al., 2001], our model give more favourable predictions.

In chapter 3, I have applied an EMT method in treating the piezoelectric problem of a 1-3 piezoelectric fiber composite. Expressions of the effective piezoelectric stress coefficients (e_{31} and e_{33}) are derived. This method has been used in treating the elastic problems of 1-3 composite and the resulting formulae shows fairly good agreement with experimental data, even at high volume fraction cases [Chen, 1998]. The formulae obtained are then combined with Chen's results [Chen, 1998] to evaluate the effective piezoelectric strain coefficients. Predictions are compared with the experimental data of d_{33} of a 1-3 PZT/epoxy composite [Klicker et. al., 1981] and the simulated values of

 e_{33} and e_{31} of a 1-3 PZT/polymer composite [Berger et. al., 2005]. Numerical EMT calculations are also carried out for comparison purposes. The d_{33} comparisons show that our predicted values agree well with the experimental data. For piezoelectric stress coefficients comparisons, there are, however, small discrepancies between the predicted values and the numerical FEM data [Berger et. al., 2005] when the volume fractions are above 0.3. This may be due to the fact that in the FEM calculation, different volume fractions were obtained by merely changing the diameter of the single fiber inside each representative volume element.

Up to now, I have used Poon and Shin's idea to find the effective properties of 0-3 piezoelectric composites. In the future, it is interesting to investigate whether this approach can be extended in treating 1-3 piezoelectric composite problems.

For the 1-3 composite, I have applied effective medium theory (EMT) to find the effective piezoelectric properties of 1-3 composites with high inclusion concentrations. However, it is applicable only for 1-3 composites having fibers with small aspect ratio. As have been mentioned in section 1.2.2, there are many theoretical works on the study of the effect of aspect ratio on the effective piezoelectric coefficients of 1-3 composites. However, those models are useful for low volume fraction composites. For further study, the idea of EMT can be extended to study the effect of aspect ratios on the effective piezoelectric coefficients of 1-3 composite at high volume fractions of the fibers.

Appendix A

Analytical elastic solutions of a single sphere composite

Goodier [Goodier, 1933] studied the elastic properties of a single inclusion embedded in an infinite matrix. Suppose a single inclusion composite was subjected to an external stress T_3 and a uniform tension T_0 is acting in the matrix far away from the inclusion. Expressions of the non-vanishing displacement (*u*) and stress (*T*) components of the constituents, in spherical coordinates (*r*, θ , φ), were found to be

$$u_{m} = -\frac{A_{1}}{r^{2}} - \frac{3A_{2}}{r^{4}} + \left[\frac{5 - 4v_{m}}{1 - 2v_{m}}\frac{A_{3}}{r^{2}} - \frac{9A_{2}}{r^{4}}\right]\cos 2\theta + \frac{T_{0}a}{2E_{m}}\left[(1 - v_{m}) + (1 + v_{m})\cos 2\theta\right]$$

$$u_{\theta m} = -\left(\frac{2A_3}{r^2} + \frac{6A_2}{r^4}\right)\sin 2\theta - \frac{T_0a}{2E_m}(1+v_m)\sin 2\theta$$

$$T_{rrm} = 2\mu_m T_0 \left\{\frac{2A}{r^3} - \frac{2v_m}{1-2v_m}\frac{C}{r^3} + \frac{12B}{r^5} + \left[-\frac{2(5-v_m)}{1-2v_m}\frac{C}{r^3} + \frac{36B}{r^5}\right]\cos 2\theta\right\}$$

$$+ \frac{T_0}{2}(1+2\cos 2\theta)$$
(A2)
(A2)

$$T_{\theta\theta m} = 2\mu_m T_0 \left[-\frac{A_1}{r^3} - \frac{2\nu_m}{1 - 2\nu_m} \frac{A_3}{r^3} - \frac{3A_2}{r^5} + \left(\frac{A_3}{r^3} - \frac{21A_2}{r^5}\right) \cos 2\theta \right] + \frac{T_0}{2} (1 - 2\cos 2\theta)$$
(A4)

$$T_{\varphi\varphi m} = 2\mu_m T_0 \left[-\frac{A_1}{r^3} - \frac{2(1-\nu_m)}{1-2\nu_m} \frac{A_3}{r^3} - \frac{9A_2}{r^5} + \left(\frac{3A_3}{r^3} - \frac{15A_2}{r^5}\right) \cos 2\theta \right]$$
(A5)

$$T_{r\theta m} = 2\mu_m T_0 \left[-\frac{2(1+v_m)}{1-2v_m} \frac{C}{r^3} + \frac{24B}{r^5} \right] \sin 2\theta - \frac{T_0}{2} \sin 2\theta$$
(A6)

$$u_{ri} = B_1 r + B_2 r + 2v_i B_3 r^3 + (3B_2 r + 6v_i B_3 r^3) \cos 2\theta$$
(A7)

$$u_{\theta i} = -[3B_2r + (7 - 4v_i)B_3r^3]\sin 2\theta$$
(A8)

$$T_{rri} = 2\mu_i T_3 \left[\frac{1 + \nu_i}{1 - 2\nu_i} B_1 + B_2 - \nu_i B_3 r^2 + (3B_2 - 3\nu_i B_3 r^2) \cos 2\theta \right]$$
(A9)

$$T_{\theta\thetai} = 2\mu_i T_3 \left\{ \frac{1+\nu_i}{1-2\nu_i} B_1 + B_2 - 5\nu_i B_3 r^2 - \left[3B_2 + 7(2-\nu_i) B_3 r^2 \right] \cos 2\theta \right\}$$
(A10)

$$T_{\varphi\varphii} = 2\mu_i T_3 \left[\frac{1+\nu_i}{1-2\nu_i} B_1 - 2B_2 - (15-7\nu_i) B_3 r^2 - (7+11\nu_i) B_3 r^2 \cos 2\theta \right]$$
(A11)

$$T_{r\theta i} = -2\mu_i T_3 \Big[3B_2 + (7 + 2\nu_i) B_3 r^2 \Big] \sin 2\theta$$
(A12)

where a, E_m , v were the radius of an inclusion, the elastic modulus of the matrix and the Poisson's ratio. A_1 , A_2 , A_3 , B_1 , B_2 and B_3 were constants. Goodier [Goodier, 1933] had worked out A_1 , A_2 and A_3 . The remaining constants can be found in Wong et. al.'s paper [Wong et. al., 2001].

$$\frac{A_{i}}{a^{3}} = -\frac{1}{8\mu_{m}} \frac{\mu_{m} - \mu_{i}}{(7 - 5\nu_{m})\mu_{m} + 2(4 - 5\nu_{m})\mu_{i}} \times \frac{2(1 - 2\nu_{i})(6 - 5\nu_{m})\mu_{m} + (3 + 19\nu_{i} - 20\nu_{m}\nu_{i})\mu_{i}}{2(1 - 2\nu_{i})\mu_{m} + (1 + 5\nu_{i})\mu_{i}} + \frac{1}{4\mu_{m}} \frac{\left[(1 - \nu_{m})\frac{(1 + \nu_{i})}{(1 + \nu_{m})} - \nu_{i}\right]\mu_{i} - (1 - 2\nu_{i})\mu_{m}}{2(1 - 2\nu_{i})\mu_{m} + (1 + \nu_{i})\mu_{i}} \tag{A13}$$

$$\frac{A_2}{a^5} = -\frac{1}{8\mu_m} \frac{\mu_m - \mu_i}{(7 - 5\nu_m)\mu_m + 2(4 - 5\nu_m)\mu_i}$$
(A14)

$$\frac{A_3}{a^3} = -\frac{1}{8\mu_m} \frac{5(1-2\nu_m)(\mu_m - \mu_i)}{(7-5\nu_m)\mu_m + 2(4-5\nu_m)\mu_i}$$
(A15)

$$B_{1} = \frac{1}{4} \frac{5(1 - v_{m})}{(7 - 5v_{m})\mu_{m} + 2(4 - 5v_{m})\mu_{i}}$$
(A16)

$$B_{2} = \frac{1}{2} \frac{(1 - \nu_{m})}{(1 + \nu_{m})} \frac{1 - 2\nu_{i}}{2(1 - 2\nu_{i})\mu_{m} + (1 + \nu_{i})\mu_{i}}$$
(A17)

$$B_3 = 0$$
 (A18)

By transforming the Goodier's solution to Cartesian coordinates, we obtain the stress components inside the constituents and their expressions are given as follows

$$T_{11i} = T_{rri} \sin^2 \theta \cos^2 \varphi + T_{\theta\theta} \cos^2 \theta \cos^2 \varphi + T_{\varphi\phi} \sin^2 \varphi + 2T_{r\theta} \sin \theta \cos \theta \cos^2 \varphi$$
$$= 2\mu_i \left(\frac{1+\nu_i}{1-2\nu_i}B_1 - 2B_2\right) = 3k_i B_1 - 4\mu_i B_2$$
(A19)

 $T_{22i} = T_{rri} \sin^2 \theta \sin^2 \varphi + T_{\theta \theta} \cos^2 \theta \sin^2 \varphi + T_{\varphi \phi i} \cos^2 \varphi + 2T_{r\theta i} \sin \theta \cos \theta \sin^2 \varphi$

$$= 2\mu_i \left(\frac{1+\nu_i}{1-2\nu_i}B_1 - 2B_2\right) = 3k_i B_1 - 4\mu_i B_2$$
(A20)

 $T_{33i} = T_{rri} \cos^2 \theta + T_{\theta\theta} \sin^2 \theta + 2T_{r\theta} \sin \theta \cos \theta$

$$= 2\mu_i \left(\frac{1+\nu_i}{1-2\nu_i}B_1 + 4B_2\right) = 3k_i B_1 + 8\mu_i B_2$$
(A21)

The general stress-strain formulae are

$$T_{11} = 2\mu \left[\frac{\nu}{1 - 2\nu} (S_{11} + S_{22} + S_{33}) + S_{11} \right]$$
$$T_{22} = 2\mu \left[\frac{\nu}{1 - 2\nu} (S_{11} + S_{22} + S_{33}) + S_{22} \right]$$

$$T_{33} = 2\mu \left[\frac{\nu}{1 - 2\nu} \left(S_{11} + S_{22} + S_{33} \right) + S_{33} \right]$$
(A22)

Using equations (A19)-(A22), expressions of strain components S of the inclusion can be obtained.

$$S_{11i} = S_{22i} = B_1 - 2B_2 \tag{A23}$$

$$S_{33i} = B_1 + 4B_2 \tag{A24}$$

For the matrix phase, its stress components were given by

$$T_{11m} = T_{22m} = 0 \tag{A25}$$

$$T_{33m} = T_0 \tag{A26}$$

Using the equation (A22), (A25) and (A26), expressions of strain components of the matrix can be obtained.

$$S_{11m} = S_{22m} = \frac{\frac{2\mu_m}{3} - k_m}{\frac{6k_m\mu_m}{6}T_0}$$
(A27)

$$S_{33m} = \frac{2(k_m + \frac{\mu_m}{3})}{6k_m \mu_m} T_0$$
(A28)

Appendix B

Derivations of the effective bulk and shear modulus for 0-3 composites having high volume fractions of the inclusions

Equation (2.19) can be written in the following form

$$\langle T_i \rangle = \langle T_m \rangle + M_1 \langle S_i \rangle - N_1 \langle S_m \rangle$$
 (B1)

where

$$\begin{split} \left\langle T_{p} \right\rangle &= \begin{pmatrix} \left\langle T_{11p} \right\rangle \\ \left\langle T_{22p} \right\rangle \\ \left\langle T_{33p} \right\rangle \end{pmatrix}, \quad \left\langle S_{p} \right\rangle &= \begin{pmatrix} \left\langle S_{11p} \right\rangle \\ \left\langle S_{22p} \right\rangle \\ \left\langle S_{33p} \right\rangle \end{pmatrix}, \\ M_{1} &= \begin{pmatrix} A + \phi C & B + \phi G & B + \phi G \\ B + \phi G & A + \phi C & B + \phi G \\ B + \phi G & B + \phi G & A + \phi C \end{pmatrix}, \quad N_{1} &= \begin{pmatrix} A & B & B \\ B & A & B \\ B & B & A \end{pmatrix} \end{split}$$

and the subscript p denotes i or m.

Since both phases are isotropic materials, their elastic behaviours can be described by equation (2.14). Using equation (2.14), (B1) can be expressed as follows

$$(R_i - M_1) \langle S_i \rangle = (R_m - N_1) \langle S_m \rangle$$
(B2)

where
$$R_p = \begin{pmatrix} k_p + \frac{4}{3}\mu_p & k_p - \frac{2}{3}\mu_p & k_p - \frac{2}{3}\mu_p \\ k_p - \frac{2}{3}\mu_p & k_p + \frac{4}{3}\mu_p & k_p - \frac{2}{3}\mu_p \\ k_p - \frac{2}{3}\mu_p & k_p - \frac{2}{3}\mu_p & k_p + \frac{4}{3}\mu_p \end{pmatrix}$$

Combining the equations of δT_{m1} , δT_{m2} and δT_{m3} , we obtain

$$(R - R_m) \langle S \rangle = \phi(R_i - R_m) \langle S_i \rangle$$
(B3)

where

$$\langle S \rangle \equiv \begin{pmatrix} \langle S_{11} \rangle \\ \langle S_{22} \rangle \\ \langle S_{33} \rangle \end{pmatrix}, \quad R = \begin{pmatrix} k + \frac{4}{3}\mu & k - \frac{2}{3}\mu & k - \frac{2}{3}\mu \\ k - \frac{2}{3}\mu & k + \frac{4}{3}\mu & k - \frac{2}{3}\mu \\ k - \frac{2}{3}\mu & k - \frac{2}{3}\mu & k + \frac{4}{3}\mu \end{pmatrix}$$

From equation (B2), we get

$$(R_{iA} + 2R_{iB})(\langle S_{11i} \rangle + \langle S_{22i} \rangle + \langle S_{33i} \rangle) = (R_{mA} + 2R_{mB})(\langle S_{11m} \rangle + \langle S_{22m} \rangle + \langle S_{33m} \rangle)$$

(B4)

$$(R_{iA} - R_{iB})(\langle S_{11i} \rangle - \langle S_{22i} \rangle) = (R_{mA} - R_{mB})(\langle S_{11m} \rangle - \langle S_{22m} \rangle)$$
(B5)

where

$$R_{iA} = k_i + \frac{4}{3}\mu_i - (A + \phi C), \quad R_{iB} = k_i - \frac{2}{3}\mu_i - (B + \phi G)$$
(B6)

$$R_{mA} = k_m + \frac{4}{3}\mu_m - A, \qquad R_{mB} = k_m - \frac{2}{3}\mu_m - B$$
 (B7)

Using equation (B3), we obtain

$$(R_{A}^{'}+2R_{B}^{'})(\langle S_{11}\rangle+\langle S_{22}\rangle+\langle S_{33}\rangle)=\phi(R_{iA}^{'}+2R_{iB}^{'})(\langle S_{11i}\rangle+\langle S_{22i}\rangle+\langle S_{33i}\rangle)$$
(B8)

$$(R_{A}^{'} - R_{B}^{'})(\langle S_{11i} \rangle - \langle S_{22i} \rangle) = \phi(R_{iA}^{'} - R_{iB}^{'})(\langle S_{11i} \rangle - \langle S_{22i} \rangle)$$
(B9)

where

$$R_{A}^{'} = (k - k_{m}) + \frac{4}{3}(\mu - \mu_{m}), \quad R_{B}^{'} = (k - k_{m}) - \frac{2}{3}(\mu - \mu_{m})$$
 (B10)

$$\vec{R}_{iA} = (k_i - k_m) + \frac{4}{3}(\mu_i - \mu_m), \quad \vec{R}_{iB} = (k_i - k_m) - \frac{2}{3}(\mu_i - \mu_m)$$
(B11)

From the equation (2.21), we have

$$(\langle S_{11} \rangle + \langle S_{22} \rangle + \langle S_{33} \rangle) = \phi(\langle S_{11i} \rangle + \langle S_{22i} \rangle + \langle S_{33i} \rangle) + (1 - \phi)(\langle S_{11m} \rangle + \langle S_{22m} \rangle + \langle S_{33m} \rangle)$$

(B12)

$$(\langle S_{11} \rangle - \langle S_{22} \rangle) = \phi(\langle S_{11i} \rangle - \langle S_{22i} \rangle) + (1 - \phi)(\langle S_{11m} \rangle - \langle S_{22m} \rangle)$$
(B13)

Substituting the equations (B4) and (B12) into equation (B8), we obtain

$$(R_{A}^{'}+2R_{B}^{'}) = \phi(R_{iA}^{'}+2R_{iB}^{'})\frac{1}{\phi+(1-\phi)\frac{(R_{iA}+2R_{iB})}{(R_{mA}+2R_{mB})}}$$
(B14)

Substituting the equations (B5) and (B13) into equation (B9), we get

$$(R_{A}^{'} - R_{B}^{'}) = \phi(R_{iA}^{'} - R_{iB}^{'}) \frac{1}{\phi + (1 - \phi)\frac{(R_{iA} - R_{iB})}{(R_{mA} - R_{mB})}}$$
(B15)

Substituting the equations (B6), (B7), (B10), (B11) and (2.12) into equations (B14) and (B15), two explicit formulae of the effective bulk modulus k and the effective shear modulus μ are obtained.

$$k = k_m + \frac{\phi(k_i - k_m)(k_m + \frac{4}{3}\mu_m)}{(1 - \phi)[k_m + \frac{4}{3}\mu_m + (1 - \phi)(k_i - k_m)] + \phi(k_m + \frac{4}{3}\mu_m)}$$

(B16)

$$\mu = \mu_m + \frac{\phi(\mu_i - \mu_m) \frac{5\mu_m (3k_m + 4\mu_m)}{6(k_m + 2\mu_m)}}{(1 - \phi)[\frac{5\mu_m (3k_m + 4\mu_m)}{6(k_m + 2\mu_m)} + (1 - \phi)(\mu_i - \mu_m)] + \phi \frac{5\mu_m (3k_m + 4\mu_m)}{6(k_m + 2\mu_m)}}{(B17)}$$

Appendix C

Effective piezoelectric coefficients of a single piezoelectric fiber composite

Consider a composite consisted of a single piezoelectric fiber inclusion embedded in a piezoelectric matrix. Both phases are transversely isotropic materials. An external electric field E is applied in its axial direction (Zdirection). We assumed that the electric field strengths inside both phases are equal to the applied electric field (i.e. $E_i = E_m = E$). Suppose the composite is poled along Z direction. The piezoelectric relations of the constituents, in our case, are given by

$$T_{rrp} = C_{11p} S_{rrp} + C_{12p} S_{\theta\theta p} + C_{13p} S_{33p} - e_{31p} E$$
(C1)

$$T_{\theta\thetap} = C_{12p} S_{rrp} + C_{11p} S_{\theta\thetap} + C_{13p} S_{33p} - e_{31p} E$$
(C2)

$$T_{33p} = C_{13p}S_{rrp} + C_{13p}S_{\theta\theta p} + C_{33p}S_{33p} - e_{33p}E$$
(C3)

$$D_{3p} = e_{31p} S_{rrp} + e_{31p} S_{\theta\thetap} + e_{33p} S_{33p} + \varepsilon_{33p} E$$
(C4)

where T, S, $C_{\alpha\beta}$, and $\varepsilon_{\alpha\beta}$ are the stress, strain, stiffness constants and dielectric constants, respectively. The subscripts r and θ represent the radial and tangential directions respectively.

The radial displacement U_{rp} and the axial displacement U_{3p} are given by

$$U_{rp} = A_p r + \frac{B_p}{r}$$
(C5)

$$U_{3p} = \xi \quad z \tag{C6}$$

where A_p , B_p and ξ are constants to be determined by boundary conditions, as follows

- (i) Along the symmetry axis of the inclusion (i.e. r = 0), the solution must be bounded. Hence $B_i = 0$.
- (ii) At the interface between the inclusion and the matrix (i.e. r = R, where
 - *R* is the radius of the fiber), we have

$$U_{ri} = U_{rm} \tag{C7}$$

$$T_{rri} = T_{rrm} \tag{C8}$$

From (C7), we get

$$A_i = A_m + \frac{B_m}{R^2} \tag{C9}$$

Before applying the boundary condition (C8), we need to determine the strain components of the constituents. Using equations (C5) and (C6), strain values can be determined.

$$S_{rrp} = A_p - \frac{B_p}{r^2} \tag{C10}$$

$$S_{\theta\theta p} = A_p + \frac{B_p}{r^2} \tag{C11}$$

$$S_{33p} = \xi \tag{C12}$$

Substituting equations (C10) to (C12) into equation (C1), we have

$$T_{rri} = C_{11i}A_i + C_{12i}S_i + C_{13i}\xi - e_{31i}E$$
(C13)

$$T_{rrm} = C_{11m} (A_m - \frac{B_m}{R^2}) + C_{12m} (A_m + \frac{B_m}{R^2}) + C_{13m} \xi - e_{31m} E$$
(C14)

Substituting equations (C9) into (C13) and (C14) and using equation (C8), we

get

$$A_{i} = \frac{2C_{11m}A_{m} + (C_{13m} - C_{13i})\xi + (e_{31i} - e_{31m})E}{C}$$
(C15)

where $C \equiv C_{11i} + C_{12i} + C_{11m} - C_{12m}$

After transforming the strain components of the constituents into Cartesian coordinates, we get

$$S_{11i} = S_{rri} \cos^2 \theta + S_{\theta \theta} \sin^2 \theta = A_i$$
(C16)

$$S_{22i} = S_{rri} \sin^2 \theta + S_{\theta\theta} \cos^2 \theta = A_i$$
(C17)

$$S_{11m} = S_{rrm} \cos^2 \theta + S_{\theta \theta m} \sin^2 \theta$$
$$= A_m - \frac{B_m}{r^2} \cos 2\theta$$
(C18)

$$S_{22m} = S_{rrm} \sin^2 \theta + S_{\theta\theta m} \cos^2 \theta$$
$$= A_m + \frac{B_m}{r^2} \cos 2\theta$$
(C19)

After taking the volumetric average, we obtain

$$\langle S_{11i} \rangle = \langle S_{22i} \rangle = A_i \text{ and } \langle S_{11m} \rangle = \langle S_{22m} \rangle = A_m$$
 (C20)

The volumetric average of the Cartesian stresses and electric displacements of both phases can then be obtained.

$$\langle T_{11p} \rangle = C_{11p} \langle S_{11p} \rangle + C_{12p} \langle S_{22p} \rangle + C_{13p} \langle S_{33p} \rangle - e_{31p} E$$
 (C21)

$$\langle T_{22p} \rangle = C_{12p} \langle S_{11p} \rangle + C_{11p} \langle S_{22p} \rangle + C_{13p} \langle S_{33p} \rangle - e_{31p} E$$
 (C22)

$$\langle T_{33p} \rangle = C_{13p} \langle S_{11p} \rangle + C_{13p} \langle S_{22p} \rangle + C_{33p} \langle S_{33p} \rangle - e_{33p} E$$
 (C23)

$$\left\langle D_{3p}\right\rangle = e_{31p}\left\langle S_{11p}\right\rangle + e_{31p}\left\langle S_{22p}\right\rangle + e_{33p}\left\langle S_{33p}\right\rangle + \varepsilon_{33p}E$$
(C24)

And the strains of the composite are obtained as follow

$$\left\langle S_{11} \right\rangle = \left\langle S_{22} \right\rangle = \phi A_i + (1 - \phi) A_m \tag{C25}$$

$$\langle S_{33} \rangle = \phi \langle S_{33i} \rangle + (1 - \phi) \langle S_{33m} \rangle = \xi$$
 (C26)

where $\langle x \rangle$ denotes the volumetric average of the physical quantity x over the respective material and ϕ is the volume fraction.

Using equation (C15) and (C25), we can obtain the following relations.

$$A_{m} = \frac{\langle S_{11} \rangle - \phi \frac{C_{13m} - C_{13i}}{C} \xi - \phi \frac{e_{31i} - e_{31m}}{C} E}{\phi \frac{2C_{11m}}{C} + 1 - \phi}$$
(C27)

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$$A_{m} = \frac{\langle S_{22} \rangle - \phi \frac{C_{13m} - C_{13i}}{C} \xi - \phi \frac{e_{31i} - e_{31m}}{C} E}{\phi \frac{2C_{11m}}{C} + 1 - \phi}$$
(C28)

From the definition of the volumetric average, we have

$$\left\langle T_{11} \right\rangle = \phi \left\langle T_{11i} \right\rangle + (1 - \phi) \left\langle T_{11m} \right\rangle \tag{C29}$$

$$\langle T_{33} \rangle = \phi \langle T_{33i} \rangle + (1 - \phi) \langle T_{33m} \rangle \tag{C30}$$

Substituting equation (C21) into equation (C29), we get

$$\langle T_{11} \rangle = \phi \Big(C_{11i} \langle S_{11i} \rangle + C_{12i} \langle S_{22i} \rangle + C_{13i} \langle S_{33i} \rangle - e_{31i} E \Big)$$

$$+ \Big(1 - \phi \Big) \Big(C_{11m} \langle S_{11m} \rangle + C_{12m} \langle S_{22m} \rangle + C_{13m} \langle S_{33m} \rangle - e_{31m} E \Big)$$
 (C31)

Substituting equation (C23) into equation (C30), we get

$$\langle T_{33} \rangle = \phi \Big(C_{13i} \langle S_{11i} \rangle + C_{13i} \langle S_{22i} \rangle + C_{33i} \langle S_{33i} \rangle - e_{33i} E \Big)$$

+ $(1 - \phi) \Big(C_{13m} \langle S_{11m} \rangle + C_{13m} \langle S_{22m} \rangle + C_{33m} \langle S_{33m} \rangle - e_{33m} E \Big)$ (C32)

Substituting equation (C20) and equation (C12) into equation (C31) and equation (C32), we obtain

$$\langle T_{11} \rangle = \phi (C_{11i}A_i + C_{12i}A_i + C_{13i}\xi - e_{31i}E) + (1 - \phi) (C_{11m}A_m + C_{12m}A_m + C_{13m}\xi - e_{31m}E)$$
(C33)

$$\langle T_{33} \rangle = \phi (C_{13i} A_i + C_{13i} A_i + C_{33i} \xi - e_{33i} E) + (1 - \phi) (C_{13m} A_m + C_{13m} A_m + C_{33m} \xi - e_{33m} E)$$
(C34)

Using equations (C15) and (C26) to (C28), $\langle T_{11} \rangle$ and $\langle T_{33} \rangle$ can be expressed in terms of $\langle S_{11} \rangle$, $\langle S_{22} \rangle$, $\langle S_{33} \rangle$ and E,

$$\langle T_{11} \rangle = C_{11} \langle S_{11} \rangle + C_{12} \langle S_{22} \rangle + C_{13} \langle S_{33} \rangle - e_{31} E$$
 (C35)

$$\langle T_{33} \rangle = C_{13} \langle S_{11} \rangle + C_{13} \langle S_{22} \rangle + C_{33} \langle S_{33} \rangle - e_{33} E$$
 (C36)

where C_{11} , C_{12} , C_{13} , C_{33} , e_{31} and e_{33} are the effective coefficients. They are now derived to be

$$C_{11} = \frac{\phi C_{11i} \frac{2C_{11m}}{C} + (1 - \phi)C_{11m}}{\phi \frac{2C_{11m}}{C} + 1 - \phi}$$
(C37)

$$C_{12} = \frac{\phi C_{12i} \frac{2C_{11m}}{C} + (1 - \phi)C_{12m}}{\phi \frac{2C_{11m}}{C} + 1 - \phi}$$
(C38)

$$C_{13} = \frac{\phi C_{13i} \frac{2C_{11m}}{C} + (1-\phi)C_{13m}}{\phi \frac{2C_{11m}}{C} + 1-\phi}$$
(C39)

$$C_{33} = \phi C_{33i} + (1 - \phi)C_{33m} + 2\phi C_{13i} \frac{C_{13m} - C_{13i}}{C}$$
$$-\frac{2\left[\phi C_{13i} \frac{2C_{11m}}{C} + (1 - \phi)C_{13m}\right]}{\phi \frac{2C_{11m}}{C} + 1 - \phi} \phi \frac{C_{13m} - C_{13i}}{C}$$
(C40)

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$$e_{31} = \frac{\phi e_{31i} \frac{2C_{11m}}{C} + (1-\phi)e_{31m}}{\phi \frac{2C_{11m}}{C} + 1 - \phi}$$
(C41)

$$e_{33} = \phi e_{33i} + (1 - \phi)e_{33m} + 2\frac{C_{13m} - C_{13i}}{C}\frac{\phi(1 - \phi)}{\phi\frac{2C_{11m}}{C} + 1 - \phi}(e_{31i} - e_{31m})$$
(C42)

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