J Mater Sci (2007) 42:6011–6017
DOI 10.1007/s10853-006-0984-9

Theoretical predictions on the effective piezoelectric coefficients of 0–3 PZT/Polymer composites

Ying Ming Poon · Chi Hin Ho · Yuen Wah Wong · Franklin G. Shin

Received: 10 April 2006 / Accepted: 21 September 2006 / Published online: 6 April 2007

Abstract Two explicit formulae for the effective piezoelectric coefficients (d_{31} and d_{33}) of 0–3 composite have been derived by taking into account the interaction effects between the inclusions and are expressed directly in terms of the properties of the constituents. Predictions using these two formulae are compared with published experimental data of 0–3 composite systems having three different polarization states: only the inclusion phase is polarized; both phases are polarized in the same direction and the two phases are polarized in opposite directions. In addition, the predictions using Wong et al.'s scheme (Wong CK, Poon YM, Shin FG (2001) J Appl Phys 90:4690) and Furukawa et al.'s model (Furukawa T, Fujino K, Fukada E (1976) Jpn J Appl Phys 15:2119) are included for comparison. For the first two cases, the results show that both our model and Wong et al.'s scheme have comparable performance. However, for the last case, our model gives more favorable predictions than theirs.

Introduction

Binary piezoelectric composites are important engineering materials and have been widely used in ultrasonic applications such as ultrasonic transducers and hydrophones [1, 2]. For a binary composite, there are 10 possible connectivity patterns [3]. Among them, 0–3 is the most common type. In the past, many theoretical models have been proposed for predictions of effective properties of 0–3 piezocomposites but they are usually applicable in dilute cases only [4–8]. Recently, Wong et al. [9] has derived two explicit piezoelectric expressions for the prediction of piezoelectric coefficients of 0–3 composites. For low volume fractions of the inclusions, they have adopted, in their scheme, the Maxwell–Wagner formula, the bulk modulus and the lower bound of the shear modulus of Hashin's model [10] for the effective dielectric constants, the effective bulk modulus and the effective shear modulus of the composite, respectively. For high volume fractions, the Bruggeman's model [11] was used to replace the Maxwell–Wagner formula for the effective dielectric constant. For the effective mechanical coefficients, the bulk modulus and the two bounds of the shear modulus of Hashin's model are used. Glushanin and Topolov [12] studied the electromechanical properties of 0–3 composite having two different structures, namely, cellular structure and rod-like structure. Taking the effects of the arrangement of the inclusions and the electromechanical interaction between two different structures, the effective elastic, piezoelectric and dielectric properties of a 0–3 composite were determined based on the effective field method. Glushanin et al. [13] took the aspect ratio of the inclusions into account and used the effective field method to determine the piezoelectric coefficients of 0–3 ($PbTiO_3$ type) ceramic/polymer composites.

In this article, we assume both phases are dielectrically and mechanically isotropic. Two explicit formulae of effective piezoelectric coefficients (d_{31} and d_{33}) for 0–3

Y. M. Poon (✉) · C. H. Ho
Department of Applied Physics and Material Research Centre, The Hong Kong Polytechnic University, Hong Kong, P.R. China
e-mail: apaympoo@polyu.edu.hk

Y. W. Wong · F. G. Shin
Centre for Smart Materials, The Hong Kong Polytechnic University, Hong Kong, P.R. China

composite are derived in terms of the electrica1, mechanical and piezoelectric coefficients of the constituents.

The following is the structure of this article. In the theory section, we extend the idea in Poon and Shin's article [14] to treat the piezoelectric problem. Expressions for the four effective stiffness constants, the two effective piezoelectric coefficients and the effective dielectric constant are derived. The results section gives comparisons of our model, Wong et. al.'s scheme, Furukawa et al.'s model [4] and the experimental data reported in the literature. The last section is the conclusion section.

Theory

In the following, we extend the idea used in Poon and Shin [14] for treating piezoelectric problems of 0–3 composites. We 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.

We shall consider a composite with volume fraction φ of the piezoelectric inclusions subjected to external stresses T_1, T_2 and T_3, and with an electric field E_3 applied in the Z direction. We further assume that both phases are dielectrically and elastically isotropic and piezoelectrically transversely isotropic.

For electric properties, Poon and Shin [14] derived the relation between the volumetric averaged electric fields inside the inclusions $\langle E_{i3} \rangle$, the matrix $\langle E_{m3} \rangle$ and the corresponding volumetric averaged electric displacements $\langle D_{i3} \rangle$ and $\langle D_{m3} \rangle$:

$$\Delta D = -2\varepsilon_m \Delta E + \phi(\varepsilon_i - \varepsilon_m)\langle E_{i3} \rangle \tag{1}$$

where $\Delta D \equiv \langle D_{i3} \rangle - \langle D_{m3} \rangle$, $\Delta E \equiv \langle E_{i3} \rangle - \langle E_{m3} \rangle$ and ε_i and ε_m are the permittivities of the inclusion and the matrix, respectively.

For piezoelectric properties, because the applied electric field can induce strain inside the constituents, Eq. 1 should be modified to

$$\begin{aligned}\Delta D = & -2\varepsilon_m \Delta E + \phi(\varepsilon_i - \varepsilon_m)\langle E_{i3} \rangle + \phi(e_{31i} - e_{31m})\langle S_{i1} \rangle \\ & + \phi(e_{31i} - e_{31m})\langle S_{i2} \rangle + \phi(e_{33i} - e_{33m})\langle S_{i3} \rangle\end{aligned} \tag{2}$$

where $\langle S_{ij} \rangle (j = 1,2,3)$ are the volumetric averaged strains in the inclusion phase, and e_{31p} and e_{33p} are the transverse and the longitudinal piezoelectric coefficients, respectively.

The above relation has assumed that both phases remain dielectrically isotropic even when they are polarized and their dielectric behavior can be described by a single parameter (ε_p).

For mechanical properties, based on Goodier's solution [15], it has been shown that [16]

$$\begin{pmatrix} \Delta T_1 \\ \Delta T_2 \\ \Delta T_3 \end{pmatrix} = \begin{pmatrix} A & B & B \\ B & A & B \\ B & B & A \end{pmatrix} \begin{pmatrix} \Delta S_1 \\ \Delta S_2 \\ \Delta S_3 \end{pmatrix} + \begin{pmatrix} \delta T_{m1} \\ \delta T_{m2} \\ \delta T_{m3} \end{pmatrix} \tag{3}$$

where $\Delta T_j \equiv \langle T_{ij} \rangle - \langle T_{mj} \rangle$, $\Delta S_j \equiv \langle S_{ij} \rangle - \langle S_{mj} \rangle (j = 1,2,3)$; $\langle T_{ij} \rangle$ and $\langle T_{mj} \rangle$ are the volumetric averaged stresses in the inclusion phase and matrix phase, respectively; $\langle S_{mj} \rangle$ are the volumetric averaged strains in the matrix phase; $A \equiv \frac{10}{9}\mu_m(-3 + \frac{2\mu_m}{k_m + 2\mu_m})$, $B \equiv \frac{1}{9}\mu_m(-3 - \frac{10\mu_m}{k_m + 2\mu_m})$; k_m and μ_m are the bulk modulus and shear modulus of the matrix, respectively;

$$\delta T_{m1} = \phi(\Delta C_{11}\langle S_{i1} \rangle + \Delta C_{12}\langle S_{i2} \rangle + \Delta C_{12}\langle S_{i3} \rangle) \tag{4}$$

$$\delta T_{m2} = \phi(\Delta C_{12}\langle S_{i1} \rangle + \Delta C_{11}\langle S_{i2} \rangle + \Delta C_{12}\langle S_{i3} \rangle) \tag{5}$$

$$\delta T_{m3} = \phi(\Delta C_{12}\langle S_{i1} \rangle + \Delta C_{12}\langle S_{i2} \rangle + \Delta C_{11}\langle S_{i3} \rangle) \tag{6}$$

where $\Delta C_{11} \equiv C_{11i} - C_{11m}$, $\Delta C_{12} \equiv C_{12i} - C_{12m}$;

$$C_{11p} = k_p + \frac{4}{3}\mu_p, \quad C_{12p} = k_p - \frac{2}{3}\mu_p;$$

k_i and μ_i are the bulk modulus and shear modulus of the inclusion, respectively.

Similar to the dielectric problem, we assume that both phases remain elastically isotropic even when they are polarized. When external stresses are applied to the piezocomposite, electric fields are induced inside the constituents. Equations 4–6 should be modified to the following:

$$\delta T_{m1} = \phi(\Delta C_{11}\langle S_{i1} \rangle + \Delta C_{12}\langle S_{i2} \rangle + \Delta C_{12}\langle S_{i3} \rangle - \Delta e_{31}\langle E_{i3} \rangle) \tag{7}$$

$$\delta T_{m2} = \phi(\Delta C_{12}\langle S_{i1} \rangle + \Delta C_{11}\langle S_{i2} \rangle + \Delta C_{12}\langle S_{i3} \rangle - \Delta e_{31}\langle E_{i3} \rangle) \tag{8}$$

$$\delta T_{m3} = \phi(\Delta C_{12}\langle S_{i1} \rangle + \Delta C_{12}\langle S_{i2} \rangle + \Delta C_{11}\langle S_{i3} \rangle - \Delta e_{33}\langle E_{i3} \rangle) \tag{9}$$

where $\Delta e_{31} \equiv e_{31i} - e_{31m}$, $\Delta e_{33} \equiv e_{33i} - e_{33m}$.

Combining Eqs. 2, 3, and 7–9, we have

$$\begin{pmatrix} \Delta T_1 \\ \Delta T_2 \\ \Delta T_3 \\ \Delta D_3 \end{pmatrix} = \begin{pmatrix} A & B & B & 0 \\ B & A & B & 0 \\ B & B & A & 0 \\ 0 & 0 & 0 & -2\varepsilon_m \end{pmatrix} \begin{pmatrix} \Delta S_1 \\ \Delta S_2 \\ \Delta S_3 \\ \Delta E_3 \end{pmatrix} + \phi \begin{pmatrix} \Delta C_{11} & \Delta C_{12} & \Delta C_{12} & -\Delta e_{31} \\ \Delta C_{12} & \Delta C_{11} & \Delta C_{12} & -\Delta e_{31} \\ \Delta C_{12} & \Delta C_{12} & \Delta C_{11} & -\Delta e_{33} \\ \Delta e_{31} & \Delta e_{31} & \Delta e_{33} & \varepsilon_i - \varepsilon_m \end{pmatrix} \times \begin{pmatrix} \langle S_{i1} \rangle \\ \langle S_{i2} \rangle \\ \langle S_{i3} \rangle \\ \langle E_{i3} \rangle \end{pmatrix} \quad (10)$$

From the definition of the volumetric average, we have

$$\begin{pmatrix} \langle T_1 \rangle \\ \langle T_2 \rangle \\ \langle T_3 \rangle \\ \langle D_3 \rangle \end{pmatrix} = \phi \begin{pmatrix} C_{11i} & C_{12i} & C_{12i} & -e_{31i} \\ C_{12i} & C_{11i} & C_{12i} & -e_{31i} \\ C_{12i} & C_{12i} & C_{11i} & -e_{33i} \\ e_{31i} & e_{31i} & e_{33i} & \varepsilon_i \end{pmatrix} \begin{pmatrix} \langle S_{i1} \rangle \\ \langle S_{i2} \rangle \\ \langle S_{i3} \rangle \\ \langle E_{i3} \rangle \end{pmatrix} + (1-\phi) \begin{pmatrix} C_{11m} & C_{12m} & C_{12m} & -e_{31m} \\ C_{12m} & C_{11m} & C_{12m} & -e_{31m} \\ C_{12m} & C_{12m} & C_{11m} & -e_{33m} \\ e_{31m} & e_{31m} & e_{33m} & \varepsilon_m \end{pmatrix} \times \begin{pmatrix} \langle S_{m1} \rangle \\ \langle S_{m2} \rangle \\ \langle S_{m3} \rangle \\ \langle E_{m3} \rangle \end{pmatrix} \quad (11)$$

$$\begin{pmatrix} \langle S_1 \rangle \\ \langle S_2 \rangle \\ \langle S_3 \rangle \\ \langle E_3 \rangle \end{pmatrix} = \phi \begin{pmatrix} \langle S_{i1} \rangle \\ \langle S_{i2} \rangle \\ \langle S_{i3} \rangle \\ \langle E_{i3} \rangle \end{pmatrix} + (1-\phi) \begin{pmatrix} \langle S_{m1} \rangle \\ \langle S_{m2} \rangle \\ \langle S_{m3} \rangle \\ \langle E_{m3} \rangle \end{pmatrix} \quad (12)$$

The constitutive piezoelectric relations are

$$\begin{pmatrix} T_{p1} \\ T_{p2} \\ T_{p3} \\ D_{p3} \end{pmatrix} = \begin{pmatrix} C_{11p} & C_{12p} & C_{12p} & -e_{31p} \\ C_{12p} & C_{11p} & C_{12p} & -e_{31p} \\ C_{12p} & C_{12p} & C_{11p} & -e_{33p} \\ e_{31p} & e_{31p} & e_{33p} & \varepsilon_p \end{pmatrix} \begin{pmatrix} S_{p1} \\ S_{p2} \\ S_{p3} \\ E_{p3} \end{pmatrix} \quad (13)$$

Using Eq. 10 and the constitutive piezoelectric relations, we get

$$[C_i - (L + \phi M)]\langle S_i \rangle = (C_m - L)\langle S_m \rangle \quad (14)$$

where

$$C_p \equiv \begin{pmatrix} C_{11p} & C_{12p} & C_{12p} & -e_{31p} \\ C_{12p} & C_{11p} & C_{12p} & -e_{31p} \\ C_{12p} & C_{12p} & C_{11p} & -e_{33p} \\ e_{31p} & e_{31p} & e_{33p} & \varepsilon_p \end{pmatrix}; \quad \langle S_p \rangle \equiv \begin{pmatrix} \langle S_{p1} \rangle \\ \langle S_{p2} \rangle \\ \langle S_{p3} \rangle \\ \langle E_{p3} \rangle \end{pmatrix}$$

$$L \equiv \begin{pmatrix} A & B & B & 0 \\ B & A & B & 0 \\ B & B & A & 0 \\ 0 & 0 & 0 & -2\varepsilon_m \end{pmatrix};$$

$$M \equiv \begin{pmatrix} \Delta C_{11} & \Delta C_{12} & \Delta C_{12} & -\Delta e_{31} \\ \Delta C_{12} & \Delta C_{11} & \Delta C_{12} & -\Delta e_{31} \\ \Delta C_{12} & \Delta C_{12} & \Delta C_{11} & -\Delta e_{33} \\ \Delta e_{31} & \Delta e_{31} & \Delta e_{33} & \varepsilon_i - \varepsilon_m \end{pmatrix}$$

Using Eqs. 12 and 13, we get

$$\langle S_i \rangle = \{\phi(C_m - L) + (1-\phi)[C_i - (L + \phi M)]\}^{-1} (C_m - L)\langle S \rangle \quad (15)$$

$$\langle S_m \rangle = \{\phi(C_m - L) + (1-\phi)[C_i - (L + \phi M)]\}^{-1} [C_i - (L + \phi M)]\langle S \rangle \quad (16)$$

where

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

Substituting Eqs. 15 and 16 into Eq. 11, we obtain

$$\langle T \rangle = \{\phi C_i \{\phi(C_m - L) + (1-\phi)[C_i - (L + \phi M)]\}^{-1} (C_m - L) + (1-\phi) C_m \{\phi(C_m - L) + (1-\phi) [C_i - (L + \phi M)]\}^{-1} [C_i - (L + \phi M)]\}\langle S \rangle \quad (17)$$

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 Eq. 17, we get

$$\begin{pmatrix} \langle T_1 \rangle \\ \langle T_2 \rangle \\ \langle T_3 \rangle \\ \langle D_3 \rangle \end{pmatrix} = \begin{pmatrix} C_{11} & C_{12} & C_{13} & -e_{31} \\ C_{12} & C_{11} & C_{13} & -e_{31} \\ C_{13} & C_{13} & C_{33} & -e_{33} \\ e_{31} & e_{31} & e_{33} & \varepsilon \end{pmatrix} \begin{pmatrix} \langle S_1 \rangle \\ \langle S_2 \rangle \\ \langle S_3 \rangle \\ \langle E_3 \rangle \end{pmatrix} \quad (18)$$

where the effective piezoelectric and mechanical coefficients are as follows:

$$
\begin{aligned}
e_{31} =\; & \phi[I_{41i}(C_{11m} - A + C_{12m} - B) + I_{43i}(C_{12m} - B) \\
& + I_{44i}e_{31m}] + (1-\phi)[I_{41m}(C_{11i} - \phi\Delta C_{11} - A \\
& + C_{12i} - \phi\Delta C_{12} - B) + I_{43m}(C_{12i} - \phi\Delta C_{12} - B) \\
& + I_{44m}(e_{31i} - \phi_{31})]
\end{aligned} \tag{19}
$$

$$
\begin{aligned}
e_{33} =\; & \phi[2I_{41i}(C_{12m} - B) + I_{43i}(C_{11m} - A) + I_{44i}e_{33m}] \\
& + (1-\phi)[2I_{41m}(C_{12i} - \phi\Delta C_{12} - B) \\
& + I_{43m}(C_{11i} - \phi\Delta C_{11} - A) + I_{44m}(e_{33i} - \phi\Delta e_{33})]
\end{aligned} \tag{20}
$$

$$
\begin{aligned}
C_{11} =\; & \phi[I_{11i}(C_{11m} - A) + (I_{12i} + I_{13i})(C_{12m} - B) \\
& + I_{14i}e_{31m}] + (1-\phi)[I_{11m}(C_{11i} - \phi\Delta C_{11} - A) \\
& + (I_{12m} + I_{13m})(C_{12i} - \phi\Delta C_{12} - B) \\
& + I_{14m}(e_{31i} - \phi\Delta e_{31})]
\end{aligned} \tag{21}
$$

The elements I_{klp} are defined below. The subscripts k,l = 1, 2, 3 refer to the three directions.

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

where

$$
\begin{pmatrix} I_{11} & I_{12} & I_{13} & -I_{14} \\ I_{12} & I_{11} & I_{13} & -I_{14} \\ I_{13} & I_{13} & I_{33} & -I_{34} \\ I_{14} & I_{14} & I_{34} & I_{44} \end{pmatrix} \equiv \left\{ \phi \begin{pmatrix} C_{11m}-A & C_{12m}-B & C_{12m}-B & -e_{31m} \\ C_{12m}-B & C_{11m}-A & C_{12m}-B & -e_{31m} \\ C_{12m}-B & C_{12m}-B & C_{11m}-A & -e_{33m} \\ e_{31m} & e_{31m} & e_{33m} & 3\varepsilon_m \end{pmatrix} \right.
$$
$$
\left. + (1-\phi) \begin{pmatrix} C_{11i}-\phi\Delta C_{11}-A & C_{12i}-\phi\Delta C_{12}-B & C_{12i}-\phi\Delta C_{12}-B & -e_{31i}+\phi_{31} \\ C_{12i}-\phi\Delta C_{12}-B & C_{11i}-\phi\Delta C_{11}-A & C_{12i}-\phi\Delta C_{12}-B & -e_{31i}+\phi_{31} \\ C_{12i}-\phi\Delta C_{12}-B & C_{12i}-\phi\Delta C_{12}-B & C_{11i}-\phi\Delta C_{11}-A & -e_{33i}+\phi_{33} \\ e_{31i}-\phi\Delta e_{31} & e_{31i}-\phi\Delta e_{31} & e_{33i}-\phi\Delta e_{33} & \varepsilon_i + 2\varepsilon_m - \phi(\varepsilon_m - \varepsilon_i) \end{pmatrix} \right\}^{-1}
$$

$$
\begin{aligned}
C_{12} =\; & \phi[I_{12i}(C_{11m} - A) + (I_{11i} + I_{13i})(C_{12m} - B) \\
& + I_{14i}e_{31m}] + (1-\phi)[I_{12m}(C_{11i} - \phi\Delta C_{11} - A) \\
& + (I_{11m} + I_{13m})(C_{12i} - \phi\Delta C_{12} - B) \\
& + I_{14m}(e_{31i} - \phi\Delta e_{31})]
\end{aligned} \tag{22}
$$

$$
\begin{aligned}
C_{13} =\; & \phi[I_{13i}(C_{11m} - A) + (I_{11i} + I_{12i})(C_{12m} - B) \\
& + I_{14i}e_{33m}] + (1-\phi)[I_{13m}(C_{11i} - \phi\Delta C_{11} - A) \\
& + (I_{11m} + I_{12m})(C_{12i} - \phi\Delta C_{12} - B) \\
& + I_{14m}(e_{33i} - \phi\Delta e_{33})]
\end{aligned} \tag{23}
$$

$$
\begin{aligned}
C_{33} =\; & \phi[I_{33i}(C_{11m} - A) + (I_{31i} + I_{32i})(C_{12m} - B) \\
& + I_{34i}e_{33m}] + (1-\phi)[I_{33m}(C_{11i} - \phi\Delta C_{11} - A) \\
& + (I_{31m} + I_{32m})(C_{12i} - \phi\Delta C_{12} - B) \\
& + I_{34m}(e_{33i} - \phi\Delta e_{33})]
\end{aligned} \tag{24}
$$

From Eq. 18, the composite as a whole is an elastically transversely isotropic material. The two effective piezoelectric coefficients (d_{31} and d_{33}) can be expressed in terms of e_{31}, e_{33} and the four effective stiffness constants. The volumetric averaged electric displacement $\langle D_3 \rangle$ of the composite is

$$
\begin{aligned}
\langle D_3 \rangle =\; & \varepsilon_{33}^T \langle E_3 \rangle + d_{31}\langle T_1 \rangle + d_{31}\langle T_2 \rangle + d_{33}\langle T_3 \rangle \\
=\; & \varepsilon_{33}^T \langle E_3 \rangle + d_{31}(C_{11}\langle S_1 \rangle + C_{12}\langle S_2 \rangle + C_{13}\langle S_3 \rangle - e_{31}\langle E_3 \rangle) \\
& + d_{31}(C_{12}\langle S_1 \rangle + C_{11}\langle S_2 \rangle + C_{13}\langle S_3 \rangle - e_{31}\langle E_3 \rangle) \\
& + d_{33}(C_{13}\langle S_1 \rangle + C_{13}\langle S_2 \rangle + C_{33}\langle S_3 \rangle - e_{33}\langle E_3 \rangle)
\end{aligned} \tag{25}
$$

where ε_{33}^T and ε_{33}^S are the effective permittivities of the composite under constant stress and constant strain conditions, respectively.

After simplifying,

$$\begin{aligned}\langle D_3\rangle =&\left(\varepsilon_{33}^T - 2d_{31}e_{31} - d_{33}e_{33}\right)\langle E_3\rangle \\ =&(d_{31}C_{11} + d_{31}C_{12} + d_{33}C_{13})\langle S_1\rangle \\ &+ (d_{31}C_{11} + d_{31}C_{12} + d_{33}C_{13})\langle S_2\rangle \\ &+ (2d_{31}C_{13} + d_{33}C_{13})\langle S_3\rangle \end{aligned} \tag{26}$$

The electric displacement can also be expressed in term of strains, as follows

$$\langle D_3\rangle = \varepsilon_{33}^S\langle E_3\rangle + e_{31}\langle S_1\rangle + e_{31}\langle S_2\rangle + e_{33}\langle S_3\rangle \tag{27}$$

Combining Eqs. 26 and 27, one can obtain

$$e_{31} = d_{31}(C_{11} + C_{12}) + d_{33}C_{13} \tag{28}$$

$$e_{33} = 2d_{31}C_{13} + d_{33}C_{33} \tag{29}$$

From Eqs. 26 and 27, the two effective piezoelectric coefficients (d_{31} and d_{33}) can be determined, as follows

$$d_{31} = \frac{\frac{e_{31}}{C_{13}} - \frac{e_{33}}{C_{33}}}{\frac{C_{11}+C_{12}}{C_{13}} - \frac{2C_{13}}{C_{33}}} \tag{30}$$

$$d_{33} = \frac{\frac{e_{31}}{C_{11}+C_{12}} - \frac{e_{33}}{2C_{13}}}{\frac{C_{13}}{C_{11}+C_{12}} - \frac{C_{33}}{2C_{13}}} \tag{31}$$

In the following section, our model is compared with existing experimental data. In addition, predictions using Wong et al.'s scheme [9] are also included in the comparisons. They assumed that the whole composite is elastically isotropic. If we also take this assumption, the two effective piezoelectric coefficients (d_{31} and d_{33}) are:

$$d_{31} = \frac{\frac{e_{31}}{C_{12}} - \frac{e_{33}}{C_{11}}}{\frac{C_{11}+C_{12}}{C_{12}} - \frac{2C_{12}}{C_{11}}} \tag{32}$$

$$d_{33} = \frac{\frac{e_{31}}{C_{11}+C_{12}} - \frac{e_{33}}{2C_{12}}}{\frac{C_{12}}{C_{11}+C_{12}} - \frac{C_{11}}{2C_{12}}} \tag{33}$$

Results

In this section, predictions made by Wong et al. [9] and our model (with and without assuming elastic isotropy) are compared with the experimental data of Furukawa et al. [17], Chan et al. [18], Zeng et al. [19] and Venkatragavaraj et al. [20] for the d_{31} of PZT/PVDF composites (with only the ceramic phase polarized), the d_{33} of PZT/P(VDF-TrFE) composites (with the two phases polarized in the same direction), d_{31}, d_{33} of PZT/P(VDF-TrFE) composites (with the two phases polarized in opposite directions) and d_{33} of a PZT/PVDF composite, in which only the ceramic phase was polarized. The parameters of the constituents adopted in our calculations are listed in Tables 1–4.

Figure 1 gives the d_{33} comparisons of the theoretical predictions with the experimental data of Chan et al. [18]. Since the piezoelectric coefficients of PZT and P(VDF-TrFE) have opposite signs, the piezoelectric activity of the composite vanished at $\phi \approx 0.45$ when both phases are polarized in the same direction and we have to set opposite signs for the piezoelectric coefficients of the constituents. It can be seen that both our model and Wong et al.'s scheme have similar reasonable performance. The small deviation between the predicted values and the experimental data are probably due to the insufficient poling [18].

Figure 2 shows the d_{31} comparisons. Since the piezoelectric activity of the composite is contributed by the inclusion phase only, we have to set d_{33m} = 0 and d_{31m} = 0. At low volume fractions, both our model and Wong et al.'s scheme show similar performance. However, at high volume fractions, some of the experimental data are not within the bounds of Wong et al.'s scheme while our model still fits well with the experimental data. We can observe the discrepancies between prediction made by our models and experimental data. This may arise from the aggregation of PZT particles [17].

Zeng et al. [19] reported the experimental data of piezoelectric coefficients d_{31} and d_{33} of the PZT/P(VDF-TrFE) composite, with both phases polarized in the opposite directions. In other words, both phases have the same sign of piezoelectric activities. The comparisons (d_{33} and d_{31}) of the experimental data with the theoretical predictions are shown in Fig. 3. When compared with Wong

Table 1 Properties of the constituents for PZT/P(VDF-TrFE) 0–3 composite adopted in our calculations in Fig. 1. Both phases are polarized in the same direction [9]

	$\varepsilon/\varepsilon_0$	Elastic Modulus (GPa)	Poisson's ratio	$-d_{31}$ (pC/N)	d_{33} (pC/N)
PZT	1159	16.8	0.35	127.9	314.4
P(VDF-TrFE)	10.7	2.32	0.39	–15.3	–38.4

Table 2 Properties of the constituents for PZT/PVDF 0–3 composite adopted in our calculations in Fig. 2. Only the inclusion phase is polarized [21]

	$\varepsilon/\varepsilon_0$	Elastic Modulus (GPa)	Poisson's ratio	$-d_{31}$ (pC/N)	d_{33} (pC/N)
PZT	1900	36	0.3	180	450
PVDF	14	1.3	0.4	0	0

Table 3 Properties of the constituents for PZT/P(VDF-TrFE) 0–3 composite adopted in our calculations in Fig. 3. Both phases are polarized in the opposite directions [19]

	$\varepsilon/\varepsilon_0$	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

Table 4 Properties of the constituents for PZT/PVDF 0–3 composite adopted in our calculations in Fig. 4. Only the inclusion phase is polarized

	$\varepsilon/\varepsilon_0^{a}$	Elastic Modulus (GPa)	Poisson's ratio	$-d_{31}$ (pC/N)	d_{33} (pC/N)
PZT	1750	63[b]	0.3[d]	175[a]	400[a]
PVDF	12	1.3[c]	0.4[e]	0	0

[a] Reference 20

[b] APC International, Ltd

[c] Reference 5

[d] Reference 21

[e] C.K. Wong, Y.M. Poon, and F.G. Shin, J. Appl. Phys. 93 (2003) 487

et al.'s scheme, our model gives more reasonable predictions to the experimental data. The abrupt decrease in the piezoelectric coefficients at $\varphi \approx 0.5$, according to [19], may be due to the redistribution of space charges at the interface of the inclusion and the matrix. This leads to some degree of depolarization and hence piezoelectric activity decreased. Both our model and Wong et al.'s scheme have assumed that the constituents are fully polarized. This may explain the discrepancies at high volume fractions.

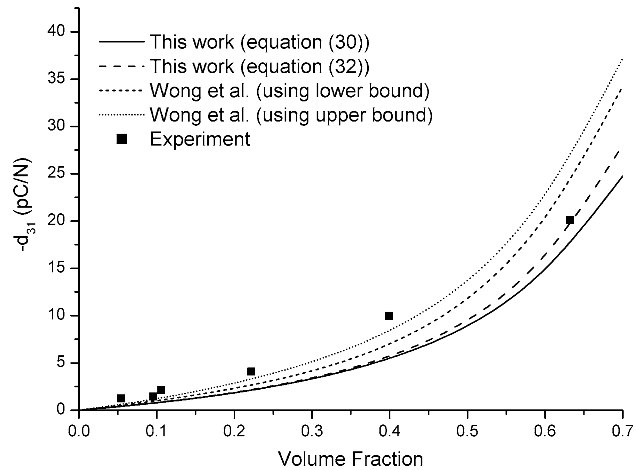

Fig. 2 Predictions of effective piezoelectric coefficient d_{31} by our model (Eqs. 30 and 32) and Wong et al.'s scheme [9] with the experimental data of Furukawa [17] of PZT/PVDF composites. Only the inclusion phase is polarized

Venkatragavaraj et al. [20] reported the experimental data of piezoelectric coefficients d_{33} of the PZT/PVDF composite prepared by two different methods; namely, hot press (HP) and solvent cast (SC) method. Since d_{33} coefficient of the composite prepared by SC method did not show a regular trend with volume fraction due to the presence of porosity and the aggregation of ceramic particles, we focus on the comparisons of the theoretical predictions by our model, Furukawa's model and Wong et al.'s scheme with the measured d_{33} of the composite prepared by HP method. They had compared their experimental results with Furukawa et al.'s model [4],

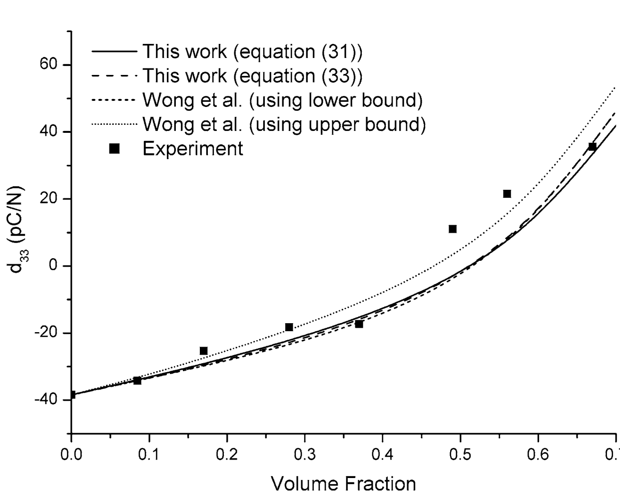

Fig. 1 Predictions of effective piezoelectric coefficient d_{33} by our model (Eqs. 31 and 33) and Wong et al.'s scheme [9] with the experimental data of Chan et al. [21] of PZT/P(VDF-TrFE) composite, with inclusion and matrix polarized in the same direction

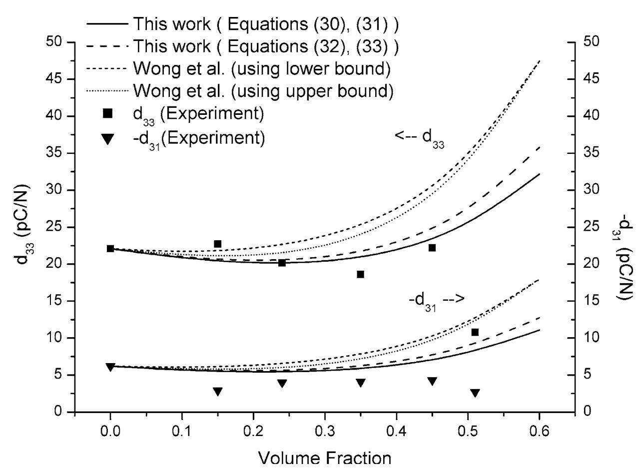

Fig. 3 Predictions of effective piezoelectric coefficients d_{33} (Eqs. 31 and 33) and d_{31} (Eqs. 30 and 32) by our model and Wong et al.'s scheme [9] with the experimental data of Zeng et al. [19] of PZT/P(VDF-TrFE) composite with inclusion and matrix polarized in opposite directions

$$d = \frac{15\phi}{(2+3\phi)(1-\phi)} \frac{\varepsilon_m}{\varepsilon_i} d_i \quad (34)$$

Equation 34 assumed that the piezoelectric activity of the composite was contributed by the inclusion phase only. In other words, Venkatragavaraj et al. have assumed that PVDF did not exhibit any piezoelectric effect. Hence as before, we took $d_{33m} = 0$ in our calculation. For the elastic modulus of the PZT APC 850, two different elastic modului (Y_{11i} = 63 GPa and Y_{33i} = 54 GPa) were provided by APC International Ltd. We have compared our d_{33} calculations by adopting two different elastic moduli. It is found that the two different sets of calculated values nearly coincide for the whole range of volume fractions of inclusion. In this comparison, we adopt Y_{11i} = 63 GPa in our calculation. Figure 4 shows that our model can give slightly better agreement with the experimental data.

From the above comparisons, it can be seen that our model (d_{31} and d_{33}), with two different assumptions concerning the mechanical properties, give similar performance. This may be due to the assumption that the constituent particulates are elastically isotropic even when they are polarized.

Conclusion

In this article, we have extended the idea of Poon and Shin [14] to obtain two explicit formulae of the two effective piezoelectric coefficients (d_{31} and d_{33}) of 0–3 composites. They are expressed directly in terms of the dielectric, elastic and piezoelectric properties of the constituents.

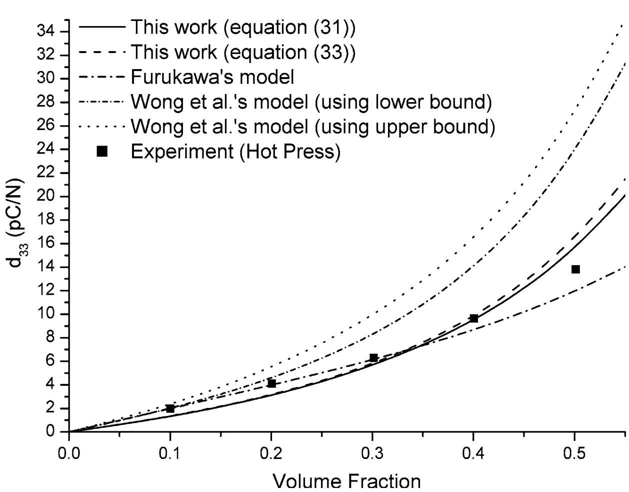

Fig. 4 Predictions of effective piezoelectric coefficient d_{33} by our model (Eqs. 31 and 33), Wong et al.'s scheme [9] and Furukawa et al.'s model [4] with the experimental data of Venkatragavaraj et al. [20] of PZT/PVDF composites prepared by hot press method (HP). Only the inclusion phase is polarized

Unlike Wong et al.'s scheme [9], our model does not need two different schemes to deal with the dilute and the concentrated suspension cases. Comparing with published experimental data of 0–3 PZT/Polymer composites, for the polarization states of having only the inclusion phase polarized or both phases polarized in the same direction, both our model and Wong et al.'s scheme give similar performance. But for the composite system with the two phases polarized in opposite directions, as compared with Wong et al.'s scheme, our model give more reasonable predictions.

Acknowledgements This work was supported by an internal research studentship (for C. H. Ho) and the Centre for Smart Materials of The Hong Kong Polytechnic University.

References

1. Ting RY, Geil FG (1991) 1990 IEEE 7th International Symposium on Applications of Ferroelectrics (Cat. No. 90CH2800–1), p 14
2. Zhou QF, Chan HLW, Choy CL (2000) Thin Solid Film 375:95
3. Newnham RE, Skinner DP, Cross LE (1978) Mater Res Bull 13:525
4. Furukawa T, Fujino K, Fukada E (1976) Jpn J Appl Phys 15:2119
5. Furukawa T, Fujino K, Fukada E (1979) J Appl Phys 50:4904
6. Yamada T, Ueda T, Kitayama T (1982) J Appl Phys 53:4328
7. Jayasundere N, Smith BV, Dunn JR (1994) J Appl Phys 76:2993
8. Prasad G, Bhimasankaram T, Suryanarayana SV, Kumar GS (1996) Modern Phys Lett B 10:517
9. Wong CK, Poon YM, Shin FG (2001) J Appl Phys 90:4690
10. Hashin Z (1962) J Appl Mech 29:143
11. Bruggeman DAG (1935) Ann Phys Lpz 24:635
12. Glushanin SV, Topolov VYu (2005) J Phys D: Appl Phys 38:2460
13. Glushanin SV, Topolov VYu, Krivoruchko AV (2006) Mater Chem Phys 97:357
14. Poon YM, Shin FG (2004) J Mater Sci 39:1277
15. Goodier JN (1933) Trans ASME 55:39
16. Ho CH, Poon YM, Wong YW, Shin FG (2006) Ferroelectric 331:1
17. Furukawa T (1989) IEEE Trans Electr Insul 24:375
18. Chan HLW, Chen Y, Choy CL (1995) Integr Ferroelectr 9:207
19. Zeng R, Kwok KW, Chan HLW, Choy CL (2002) J Appl Phys 92:2674
20. Venkatragavaraj E, Satish B, Vinod PR, Vijaya MS (2001) J Phys D: Appl Phys 34:487
21. Wong CK, Shin FG (2005) J Appl Phys 97:034111