Theoretical Study on the Static Performance of Piezoelectric Ceramic–Polymer Composites with 2–2 Connectivity

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Abstract—The static equilibrium conditions have been derived for piezoelectric-ceramic-polymer composite structures. Rigorous solutions are obtained for a 2–2 composite of lamellar configuration. Under a uniaxial stress or an electric field the strain profile becomes inhomogeneous due to different elastic stiffness of the two components (hard piezoelectric ceramic and soft polymer). The stress transfer between the two components is identified as due to an additional shear stress produced at the ceramic-polymer interface, and the amplification factor is defined for the enhancement of the response of the active piezoelectric ceramic resulting from this stress transfer. It is shown that this enhancement effect not only depends on the volume percentage of the active component but also depends on the aspect ratio of the two components.

I. INTRODUCTION

COMPOSITE piezoelectric materials are usually made of active piezoelectric ceramic (such as lead zirconate titanate (PZT)) rods or plates embedded in a passive polymer matrix. The shape of the active and passive components vary according to the application requirements. The composites are usually categorized according to their connectivity [1].

Combining the merits of both components and utilizing the interface effect in an integrated structure of hard and soft materials, the composite piezoelectric materials are proved to be an excellent new class of transducer materials for medical imaging and underwater hydrophone applications [2], [3]. The advantage of the ceramic-polymer composite structures is the adjustability of their physical properties, such as density, elastic compliance, dielectric properties, piezoelectric properties, acoustic impedance etc., through manipulating the contents and the geometries of the active and passive components.

One of the important factors for the success of a composite structure is the effectiveness of stress transfer from the passive phase to the active phase. This stress transfer effectively increases the response of the active component, so that one can reduce the volume percentage of the ceramic phase to lower the total system acoustic impedance and density without affecting the piezoelectric properties. In order to understand the stress transfer and its induced phenomena in the composite structures, several theoretical models [4]–[6] were developed in the past based on certain assumptions. Although these mod-

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II. STATIC EQUILIBRIUM CONDITIONS FOR A 2–2 COMPOSITE UNDER UNIAXIAL STRESS

The cross section of a 2–2 lamellar diphasic polymer-ceramic composite is shown in Fig. 2, where $a$ and $d$ are the dimensions of the PZT and the polymer respectively in the $x$-direction, and $l$ is the thickness of the composite in the $z$-direction. Assuming the dimensions of the composite are large in the $x$- and $y$-directions, the system may be treated as one-dimensional. For convenience, we cut a slab of width $h$ in the $y$-direction as shown in Fig. 2, which will be used for the following analyses and throughout this paper.

Under a uniaxial stress $T_3$, both the polymer and the ceramic are either stretched or compressed depending on the sign of $T_3$. For simplicity, we do not consider in this paper the inhomogeneity in the $y$-direction but only focus on the inhomogeneous displacement profile in the $z$-direction. The equilibrium conditions will be derived for the polymer phase only; one may obtain the equilibrium conditions for the ceramic phase by analogy. In the static case, the system has enough time to adjust its shape in response to a given external stress, so that we can assume the strain to be uniform in the $z$-direction for any given $x$. From symmetry consideration, the $z = 0$ plane (mirror plane) does not move at all in the $z$-direction when the system is under a uniaxial stress $T_3$ or under an electric field $E_3$. Using these two arguments one can immediately write down the following ansatz for the displacement $u(x, z)$ in the $z$-direction:

$$u(x, z) = \frac{2z}{l} u \left( x, \frac{z}{2} \right)$$

where $u(x, l/2)$ is the displacement profile at the top surface of the composite.

Let us now derive the static equilibrium condition for the polymer phase using (1). Taking a small element $h \, dx \, dz$ as
shown in Fig. 2, the shear force in the $z$-direction is
\[ h \, dz \, \mu^p \, u_{xz}(x, z) \, dx = \frac{2z}{l} \mu^p \, u_{xz}(x, \frac{l}{2}) \, h \, dz \, dx \] (2)
where $\mu^p$ is the shear modulus of the polymer. Because the displacement field is symmetric with respect to the $z = 0$ plane, we only need to consider the upper half of the composite. From (2) one can calculate the total shear force $f_s$ acting on the small slab $h \cdot dz \cdot (l/2)$ in Fig. 2:
\[ f_s = \frac{l}{2} \, \mu^p \, u_{xz}(x, \frac{l}{2}) \, h \, dx \] (3)
This force reflects the coupling of neighboring slab elements when the strain field is inhomogeneous. Another force on the slab $h \cdot dz \cdot (l/2)$ is the restoring force $f_r$ against the elongation in the $z$-direction, which is
\[ f_r = -Y^p \frac{u(x, l/2)}{l/2} \, h \, dx \] (4)
where $Y^p$ is the Young’s modulus of the polymer. The third force acting on this small slab is the external force $f_e$ resulting from the uniaxial stress $T_3$:
\[ f_e = T_3 \, h \, dx. \] (5)
In static equilibrium the total forces on this slab should be zero, i.e.,
\[ f_s + f_r + f_e = 0. \] (6)
Substituting (3)–(5) into (6) we obtain the following second order differential equation:
\[ \frac{l}{4} \mu^p u_{xx}(x, l/2) - Y^p u(x, l/2) + T_3 = 0 \] (7)
which is the static equilibrium condition for the polymer phase in terms of its surface displacement.

III. SOLUTIONS FOR DISPLACEMENT PROFILE AND EFFECTIVE PHYSICAL PROPERTIES OF A 2–2 COMPOSITE

A. Surface Displacement of the Composite

Because the solution should be symmetric with respect to the $x = 0$ plane according our choice of the coordinate system (see Fig. 2), we only keep the even solution in solving (7). The displacement profile of the polymer surface is
\[ u(x, l/2) = A \, ch \left( \frac{2}{l} \sqrt{\frac{2Y^p}{\mu^p}} \right) \, \frac{l/2}{Y^p} T_3, \quad \frac{d}{2} < x < \frac{d}{2} \] (8a)
\[ u(x + n[a + d], z) = u(x, z), \quad n = 1, 2, 3, \ldots. \] (8b)
Here $A$ is an integration constant to be determined, (8b) represents the periodic nature in the $x$-direction with a period of $(a + d)$, and $ch(x)$, $sh(x)$, $th(x)$ and $cth(x)$ used here and later in the text are the hyperbolic functions.

Following exactly the same procedure described previously, one can obtain the inhomogeneous displacement $u(x, l/2)$ of the ceramic surface in the $z$-direction:
\[ u(x, l/2) = B \, ch \left[ \frac{2}{l} \sqrt{\frac{2}{\sigma_{33}C_{44}}} \left( x - \frac{a + d}{2} \right) \right] + \frac{l}{2} \, s_{33} T_3, \quad \frac{d}{2} < x < \frac{d}{2} + a \] (9a)
\[ u(x + n[a + d], z) = u(x, z), \quad n = 1, 2, 3, \ldots. \] (9b)
where $s_{33}$ is the compliance in $z$-direction and $c_{44}$ is the shear elastic constant for the ceramic, $B$ is an integration constant to be determined.

At the present time we only deal with the situation with no interfacial slip, so that the first boundary condition is
\[ u(d/2, l/2) = u(d/2, l/2). \] (10)

Another condition may be derived from the Newton’s third law. We know that the inhomogeneous displacement profile is produced by the additional shear force acting between the polymer and the ceramic at the interface, and this additional force can be calculated from the extradosplacement. By the term extradosplacement we mean the difference of the displacement profile of the composite from the displacement of single pure phase under the same uniaxial stress. For instance, the total additional force $F_p$ experienced by the polymer in the region of $0 < x < d/2$, which is provided by the ceramic in the region of $d/2 < x < (a + d)/2$ through the the tight binding at the interface, is given by
\[ F_p = \int_0^{d/2} \frac{Y^p}{l/2} \left[ u(x, l/2) - \frac{l/2}{Y^p} T_3 \right] \, h \, dx \] (11)
\[ = A \, h \, \sqrt{\frac{\mu^p Y^p}{2}} \, sh \left( \frac{d}{l} \sqrt{\frac{2Y^p}{\mu^p}} \right) \] (11)
where the term $\left( \frac{l/2}{Y^p} T_3 \right)$ is the displacement of a single phase polymer under stress $T_3$. Similarly, the shear force experienced by the ceramic counterpart is given by
\[ F_c = B \, h \, \sqrt{\frac{C_{44}}{2\sigma_{33}}} \, sh \left( \frac{a}{l} \sqrt{\frac{2}{\sigma_{33}C_{44}}} \right). \] (12)
From Newton’s third law we have
\[ F_p = -F_c \] (13)
which together with (10) can serve as the conditions to help us determine the integration constants $A$ and $B$:
\[ A = \frac{1}{l} \left( \sigma_{33} - 1/Y^p \right) T_3 \sqrt{\frac{\mu^p Y^p s_{33}}{c_{44}}} \, sh \left( \frac{d}{l} \sqrt{\frac{Y^p}{\mu^p}} \right) cth \left( \frac{a}{l} \sqrt{\frac{2}{\sigma_{33}C_{44}}} \right) + ch \left( \frac{d}{l} \sqrt{\frac{2Y^p}{\mu^p}} \right) \] (14)
\[ B = \frac{-\frac{1}{2}(\sigma_{33} - 1/Y^P) \cdot T_3}{\sqrt{\frac{\sigma_{33}}{\mu^0 Y^P}} \cdot \frac{a h}{2}} \]

(15)

**B. Effective Piezoelectric Constants \( \bar{d}_{33} \) and \( \bar{g}_{33} \)**

The surface displacements of both polymer and ceramic (8) and (9) are homogeneous under the uniaxial stress \( T_3 \), which is caused by the stress transfer from the soft to the hard component in the composite structure. The local stress, which is now a combination of the applied normal stress and the additional shear stress, is not uniform. Hence, the electric displacement that is proportional to the local applied stress on the ceramic also becomes inhomogeneous. Because the electric displacement is equal to the surface charge density for a parallel plates capacitor, the total charge produced by the active piezoelectric ceramic in the range of \( d/2 < x < (a + d)/2 \) can be obtained from the following integration:

\[ Q = \int_{d/2}^{(a+d)/2} D(x)h \, dx = \frac{\gamma d_{33} a h}{2} T_3 \]

(16)

where \( \gamma \) is the stress amplification factor:

\[ \gamma = 1 \left( 1 + \sqrt{\frac{\frac{1}{2}(1/Y^P - s_{33})}{\mu^{0} Y^P}} \right) \frac{2\pi a}{\sqrt{\frac{2s_{33}}{c_{44}}} \cdot \frac{2\pi a}{\sqrt{\frac{2s_{33}}{c_{44}}}}} \]

(17)

which characterizes the amount of stress increase in the ceramic phase. In order to maximize \( \gamma \), from (17) one should choose a passive material with a small Young's modulus but a large shear modulus. The maximum \( \gamma \) value can be obtained in this way is

\[ \lim_{\mu^{0} Y^P \to 0} \gamma = 1 + \frac{d}{a} \]

(18)

dimensions are involved in the expression. Generally speaking, \( l \) should be large and \( a \) should be small in order to get a larger \( \gamma \). Fig. 3 shows the changes of the \( \gamma \) values for a PZT-Epoxy composite with respect to the volume percent of ceramic phase at different \( (a + d)/l \) ratio. The elastic properties used in the calculations are listed in Table I. One can see from Fig. 3 that \( \gamma \) increases with the decrease of the volume percent of ceramic or the decrease in the ratio of \( (a + d)/l \). We can derive the limit of \( \gamma \) for large \( l \) from (17):

\[ \lim_{l \to \infty} \gamma = \frac{1}{\nu^c + \mu^p Y^P s_{33}} \]

(19)

where \( \nu^c \) and \( \mu^p \) are the volume fractions of the ceramic and the polymer phases respectively. This result is identical as that obtained from the isostain approximation, which indicates that the calculated values for a composite structure using isostain approximation are the upper limits of the corresponding physical quantities.

When \( l \) is fixed, smaller \( a \) means smaller volume percentage of ceramic phase. Although the \( \gamma \) value may be increased by reducing \( a \), the total composite system might have smaller effective piezoelectric \( d_{33} \) constant due to the reduction of the active phase content. Considering the periodic nature in the \( x \)-direction and assuming the polymer to be nonpiezoelectric, then for the 2–2 composite shown in Fig. 2 the charge produced by the ceramic in the range of \( d/2 < x < (a + d)/2 \) will be distributed over the range of \( 0 < x < (a + d)/2 \) in order to maintain the equal potential surfaces at the top and bottom electrodes. Therefore, the effective piezoelectric \( d_{33} \)
The effective piezoelectric constant for the composite is
\[
\bar{d}_{33} = \frac{a}{a + d} \gamma d_{33} = v^e \gamma d_{33}
\]  
(20)

Fig. 4 are the plots of \( \bar{d}_{33} / d_{33} \) values for different ratio of \((a + d)/l\), one can see clearly that the aspect ratio of the ceramic plays a crucial role in the total piezoelectric effect of the composite system. When the ratio \((a + d)/l\) is large, the enhancement effect will be small and the \( d_{33} \) is almost linearly increasing with the volume percent of the active ceramic phase except near the two ends of the curve. As the \((a + d)/l\) ratio decreases, the enhancement effect increases but the \( d_{33} \) is always less than \( d_{33} \) so long as the ceramic content is less than 100%. The effect of the aspect ratio is more pronounced at low ceramic content.

The effective piezoelectric \( \overline{d}_{33} \) constant can be derived using the condition of equal electric field inside both the polymer and ceramic phases (result of equal potential electrode surfaces):
\[
\overline{d}_{33} = \frac{\gamma d_{33}}{\varepsilon^e + (d/a)\varepsilon^p}
\]  
(21)

where \( \varepsilon^e \) and \( \varepsilon^p \) are the dielectric constants of the ceramic and polymer respectively.

In common practice, the product \( d_{33} \overline{d}_{33} \) is often used as the figure of merit for a given composite structure, this value is plotted in Fig. 5 against the volume content of PZT for several values of \((a + d)/l\) ratio. One can see from Fig. 5 that if use \( d_{33} \overline{d}_{33} \) as the sole design criterion, the best composites are those containing around 10% active ceramic phase by volume for the composite discussed here. Note the values in Table I are for pure PZT and Spurr’s Epoxy taken from [10] and [11] respectively. If doped PZT’s are used, the absolute values in Fig. 5 will be changed because the \( d_{33} \) and \( \varepsilon^p \) values will differ greatly. However, the general trend shown in Fig. 5 is correct.

C. The Converse Piezoelectric Effect

When we apply an electric field instead of pressure to a composite, similar inhomogeneous surface profile will be obtained. The only difference is that the electric field only have effect on the piezoelectric ceramic component but not on the passive component. Therefore the solutions (8a)–(9b) are simplified to

\[
u(x, l/2) = A’ \cosh \left( \frac{2}{l} \sqrt{\frac{2Y_p}{\mu_p}} \left( x - \frac{d}{2} \right) \right) \cosh \left( \frac{2}{l} \sqrt{\frac{2Y_p}{\mu_p}} \left( x + \frac{d}{2} \right) \right)
\]  
(22a)

\[
u(x + n(a + d), z) = u(x, z), \quad n = 1, 2, 3, \cdots
\]  
(22b)

and

\[
u(x, l/2) = B’ \cosh \left( \frac{2}{l} \sqrt{\frac{2}{\sigma_{33}C_{44}}} \left( x - \frac{a + d}{2} \right) \right) + \frac{l}{2} d_{33} E_3,
\]  
(23a)

\[
u(x + n(a + d), z) = v(x, z), \quad n = 1, 2, 3, \cdots
\]  
(23b)

with \( A’ \) and \( B’ \) given in (24) and (25), respectively (shown at the bottom of the page).

For convenience, we like to define an average stain for the composite, which formally still obeys the constitutive relations. The arithmetic mean will suffice this requirement so that we define

\[
A’ = \frac{\frac{1}{2} d_{33} E_3}{\sqrt{\frac{\mu Y_p \sigma_{33}}{\epsilon_{44}}} \sinh \left( \frac{\frac{1}{2} \sqrt{\frac{2Y_p}{\mu_p}}}{\sigma_{33}C_{44}} \right) \cosh \left( \frac{\frac{1}{2} \sqrt{\frac{2Y_p}{\mu_p}}}{\sigma_{33}C_{44}} \right) + \cosh \left( \frac{\frac{1}{2} \sqrt{\frac{2Y_p}{\mu_p}}}{\sigma_{33}C_{44}} \right)}
\]  
(24)

\[
B’ = \frac{\frac{1}{2} \sigma_{33} - 1/Y_p \cdot T_3}{\sqrt{\frac{\mu Y_p \sigma_{33}}{\epsilon_{44}}} \sinh \left( \frac{\frac{1}{2} \sqrt{\frac{2Y_p}{\mu_p}}}{\sigma_{33}C_{44}} \right) \cosh \left( \frac{\frac{1}{2} \sqrt{\frac{2Y_p}{\mu_p}}}{\sigma_{33}C_{44}} \right) + \cosh \left( \frac{\frac{1}{2} \sqrt{\frac{2Y_p}{\mu_p}}}{\sigma_{33}C_{44}} \right)}
\]  
(25)
\[ \overline{S_3} = \frac{2}{(a + d)(l/2)} \left[ d/2 \int_0^{d/2} u(x,l/2) \, dx + \int_{d/2}^{(a+d)/2} v(x,l/2) \, dx \right] \]
\[ = d_{33} E_3 \tag{26} \]

where \( d_{33} \) is given by (19).

IV. CONCLUSION

A formal theory has been formulated for the ceramic–polymer composite structures, which has successfully described the inhomogeneous deformation of a 2–2 composite under a uniaxial stress and/or under a static electric field. The novel feature of the present theory is the elimination of the isostrain approximation, which enables us to include the aspect ratio of each component in the characterization of the composites.

The mechanism of the stress transfer in composite structures under a static elastic stress (electric field) can be explained as follows. Due to the difference in their elastic stiffness (piezoelectric properties), the polymer and ceramic phases should deform different amounts under a given stress (electric field). However, the nonslip interface forces the displacements of the two phases to be the same at the interface so that a shear stress is generated through this nonslip bonding. This inhomogeneous shear stress in turn produces additional nonuniform deformations in both the ceramic and the polymer phases, producing enhancement to the response of the active harder ceramic at the expense of the response of the passive softer phase (the effect is the opposite for the converse piezoelectric response).

An amplification factor \( \gamma \) has been defined, which is the ratio of the effective total stress on the ceramic and the actually applied external stress. This amplification factor \( \gamma \) characterizes the effectiveness of the stress transfer in a given composite structure.

It is found that the effective piezoelectric constant of the composite not only depends on the volume percentage of the active component, but also strongly depends on the aspect ratio and the configurations. Our theory predicts that the efficient composite structures should have a small \((a + d)/l\) ratio, and the passive material should be selected to have small Young's modulus but large shear modulus. Surface capping effectively increases the ratio of \( \mu_0 / Y_p \) in the direction normal to the capping surface, hence is a better structure according to the theory. Because the results from isostrain models provide only the upper limits of those calculated physical quantities, they are generally larger than the experimental values. In comparison, the results obtained from the current theory agree very well with the experimental results [9].

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REFERENCES


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