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Analytical modelling of 3-3 piezoelectric composites

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Abstract

A piezoelectric composite of the '3-3' type consists of interpenetrating active piezoceramic and passive polymer phases. Applications for these materials include low frequency hydrophones, due to the improved properties and figures of merit under hydrostatic conditions. Previous research on analytical modelling 3-3 materials has considered polymer infiltrated equiaxed open pores in a piezoelectric ceramic and has assumed complete stress transfer of an applied load into the stiffer ceramic matrix. This research extends the analytical modelling of 3-3 piezocomposites to include the load experienced by the polymer phase. The model is a useful aid to fabricate and optimise piezocomposite 3-3 structures for specific applications. © 2001 Elsevier Science Ltd. All rights reserved.

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1. Introduction

Piezoelectric composites have been developed in an attempt to improve on the properties of monolithic piezoelectric materials, such as lead zirconate titanate (PZT), and are of interest for applications such as acoustic transducers, medical imaging and non-destructive evaluation.[1] These materials consist of an active piezoelectric phase and a passive phase, usually a polymer. The composites of concern in this paper are classified as '3-3' composites where both the piezoelectric and polymer are connected in three dimensions. In simple terms, this can be regarded as an open porosity piezoelectric material that has been impregnated with a polymer phase.[2]

To optimise the properties of 3-3 piezocomposites, knowledge of the figures of merit that indicate the performance of the material for specific applications is required. If the material is used as a hydrophone to detect a low frequency signal (< 100 kHz), the acoustic wavelength dimension is greater than that of the hydrophone and the stress on the composite due to the acoustic wave is hydrostatic.[3] Passive hydrophone sensitivity is characterised by the *hydrostatic piezoelectric voltage constant* (g_h), defined as the electric field generated per unit hydrostatic stress,[4] which is calculated from Eq. (1).

$$g_h = \frac{d_h}{\varepsilon_{33}^T} \ (\mathrm{Vm^{-1}\ Pa^{-1}}) \qquad (1)$$

where ε_{33}^T is the permittivity at constant stress of the composite (F m^{-1}) and d_h is the hydrostatic strain constant (m V^{-1}), which is defined as the hydrostatic strain per unit electric field[4] or charge developed per unit hydrostatic force and given by;

$$d_h = d_{33} + 2d_{31} \left(\mathrm{mV^{-1}\ or\ C\,N^{-1}}\right) \qquad (2)$$

where d_{33} is the charge per unit force applied parallel to the poling direction (3-direction), d_{31} is the charge per unit force applied perpendicular to the poling direction.

Both d_h and g_h values are used to calculate the *hydrostatic figure of merit* ($d_h \times g_h$), which describes the device both as a hydrophone and an actuator (active–passive transducer) and indicates the signal to noise ratio.[4,5]

While many papers have modelled the behaviour 0-3 and 1-3 composites, there has been less research in the area of 3-3 composites. The most useful models developed by Rittenmyer et al.[6] and Banno[7] have equiaxed pores and assume that the polymer phase bears no load due to complete stress transfer of the load into the stiffer piezoelectric phase. The aim of his paper is to modify

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the 3-3 model to allow examination of how the hydrostatic properties of 3-3 piezocomposites are affected by polymer volume fraction and the elastic modulus of the polymer phase.

2. Modelling of 3-3 piezocomposites

Rittenmyer et al.[6] reported an analytical model describing the physical properties of 3-3 piezocomposites. The model consists of three intersecting rectangular columns of piezoelectric embedded in a polymer matrix, as shown in Fig. 1. This model assumes that the piezoelectric material is fully poled parallel to the poling direction (*z*- or 3-direction).

2.1. Permittivity

The permittivity at constant stress in the 3-direction ($\varepsilon_{33}^{\mathrm{T}}$) is of importance in calculating the hydrostatic voltage constant ($g_{\mathrm{h}}=d_{\mathrm{h}}/\varepsilon_{33}^{\mathrm{T}}$) of the composite. As the permittivity of the piezoelectric phase is significantly greater than that of the polymer, it will dominate the effective permittivity of the composite. By examination of Fig. 1, the column parallel to the poling direction can be considered to contribute to $\varepsilon_{33}^{\mathrm{T}}$of the composite (volume 1 in Fig. 1). The other volumes contribute little to the effective permittivity of the composite as they consist of either purely polymer (volume 2) or a piezoelectric phase in series in the 3-direction with the low permittivity polymer phase (volumes 3 and 4). For the model cube, the dielectric constant can be approximated to Eq. (3).[7]

$$\varepsilon_{33}^{\mathrm{T,Composite}}=\varepsilon_{33}^{\mathrm{T,PZT}}.\ v^{\varepsilon 33\mathrm{PZT}} \tag{3}$$

where $\varepsilon_{33}^{\mathrm{T,Composite}}$ is the effective permittivity at constant stress of the composite (F m^{-1}), $\varepsilon_{33}^{\mathrm{T,PZT}}$is the permittivity at constant stress of the piezoelectric phase (F m^{-1}) and $\mathrm{v}^{\varepsilon 33\mathrm{PZT}}=[L/(L+l_2)]^2$ is the volume fraction of piezoelectric phase contributing to the composite permittivity.

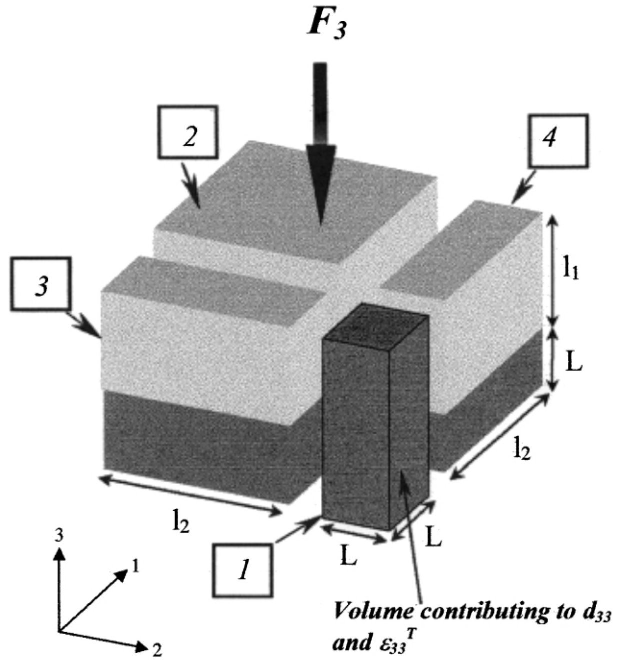

Fig. 1. Determination of d_{33}. As a force F_3 is applied, the load is distributed between volumes 1–4. The distribution depends of the respective cross-sectional areas and compliances.

2.2. Piezoelectric strain constant — d$_{33}$

In order to calculate the d_{33} piezoelectric charge coefficient of the composite material, the fraction of force experienced by the active piezoelectric material must be calculated. If a force, F_3 is applied in the 3-direction, the total force is distributed through volumes 1–4 indicated in Fig. 1. The polymer phase will experience less force, being of significantly higher compliance than the piezoceramic. This has led to the assumption in some models of complete stress transfer into the active ceramic phase.[6] However, the actual distribution of the force will depend on the respective compliances of the volumes and the cross sectional area of each volume, similar to that of composite fibre reinforced systems. For volume 1, the compliance s_{33}^{V1} is that of the piezoelectric material, defined as $s_{33}^{\mathrm{PZT}}$and the area $a^{\mathrm{V1}}=L^2$. For volume 2, the compliance s_{33}^{V2} is that of the polymer, defined as $s_{33}^{\mathrm{Polymer}}$ and the area is $a^{\mathrm{V2}}=l_2.l_2$. Volumes 3 and 4 are identical in geometry and consist of a polymer phase and a piezoelectric phase connected in series in the 3-direction. The area of both volumes is $a^{\mathrm{V3}}=a^{\mathrm{V4}}=L.l_2$ and the compliance of the volumes in the 3-direction can be calculated using a series model [3] [Eq. (4)]:

$$\begin{aligned} s_{33}^{\mathrm{V3}}=s_{33}^{\mathrm{V4}}&=v^{\mathrm{V3}}.s_{33}^{\mathrm{PZT}}+(1-v^{\mathrm{V3}}).s_{33}^{\mathrm{Polymer}}\\ &=\frac{L.s_{33}^{\mathrm{PZT}}+l_1.s_{33}^{\mathrm{Polymer}}}{(L+l_1)} \end{aligned} \tag{4}$$

where $\mathrm{v}^{\mathrm{V}3}$ is the volume fraction of piezoelectric phase in volume 3, $s_{33}^{\mathrm{V3}}$is the compliance of volume 3 (Pa^{-1}) and s_{33}^{V4} is the compliance of volume 4 (Pa^{-1}).

Volume 2 consists of a passive polymer phase only and has no active phase, while the piezoelectric phases in volumes 3 and 4 are in series with the polymer in the 3-direction, which results in a negligible contribution to d_{33}. Thus, only volume 1 can be considered to contribute to the effective d_{33} of the composite. For a composite consisting of materials with compliances s_{33}^{V1}, s_{33}^{V2}, s_{33}^{V3} and s_{33}^{V4} with areas of a^{V1}, a^{V2}, a^{V3} and a^{V4}, the ratio of force within each volume is given by $\frac{a^{\mathrm{V1}}}{s_{33}^{\mathrm{V1}}}:\frac{a^{\mathrm{V2}}}{s_{33}^{\mathrm{V2}}}:\frac{a^{\mathrm{V3}}}{s_{33}^{\mathrm{V3}}}:\frac{a^{\mathrm{V4}}}{s_{33}^{\mathrm{V4}}}$.

The d_{33} of the 3-3 piezocomposite structure $\left(d_{33}^{\mathrm{Composite}}\right)$ can therefore be calculated by multiplying d_{33} coefficient of PZT (d_{33}^{PZT}) with the fraction of load experienced by volume 1.

$$d_{33}^{\mathrm{Composite}} = d_{33}^{\mathrm{PZT}} \cdot \frac{a^{\mathrm{V1}}}{s_{33}^{\mathrm{V1}}} \cdot \left[\frac{a^{\mathrm{V1}}}{s_{33}^{\mathrm{V1}}} + \frac{a^{\mathrm{V2}}}{s_{33}^{\mathrm{V2}}} + \frac{2a^{\mathrm{V3}}}{s_{33}^{\mathrm{V3}}}\right]^{-1}$$
$$= d_{33}^{\mathrm{PZT}} \cdot \frac{L^2}{s_{33}^{\mathrm{PZT}}} \cdot \left[\frac{L^2}{s_{33}^{\mathrm{PZT}}} + \frac{l_2.l_2}{s_{33}^{\mathrm{Polymer}}} + \frac{2.L.l_2.(L+l_1)}{L.s_{33}^{\mathrm{PZT}} + l_1.s_{33}^{\mathrm{Polymer}}}\right]^{-1} \tag{5}$$

2.3. *Piezoelectric strain constant* — d$_{31}$

The same procedure is undertaken to calculate the d_{31} piezoelectric charge coefficient of the composite. If a force, F_1 is applied in the 1-direction, the total force is distributed through the volumes 5–8, indicated in Fig. 2. For volumes 5 and 6, which are single phases of polymer and piezoelectric, respectively, the compliances are $s_{11}^{\mathrm{V5}} = s_{11}^{\mathrm{Polymer}}$ and $s_{11}^{\mathrm{V6}} = s_{11}^{\mathrm{PZT}}$ and the areas are $a^{\mathrm{V5}} = l_1.l_2$ and $a^{\mathrm{V6}} = L^2$. For volumes 7 and 8, the compliance is calculated using a series model[6] [Eq. (6)]:

$$s_{11}^{\mathrm{V7}} = s_{11}^{\mathrm{V8}} = v^{\mathrm{V8}}.s_{11}^{\mathrm{PZT}} + (1 - v^{\mathrm{V8}}).s_{11}^{\mathrm{Polymer}}$$
$$= \frac{L.s_{11}^{\mathrm{PZT}} + l_2.s_{11}^{\mathrm{Polymer}}}{(L + l_2)} \tag{6}$$

where v^{V8} is the volume fraction of piezoelectric in volume 7 or 8, s_{11}^{V7} is the compliance of volume 7 (Pa^{-1}) and s_{11}^{V8} is the compliance of volume 8 (Pa^{-1}). The area of volumes 7 and 8 are given by $a^{\mathrm{V7}} = L.l_1$ and $a^{\mathrm{V8}} = L.l_2$, respectively.

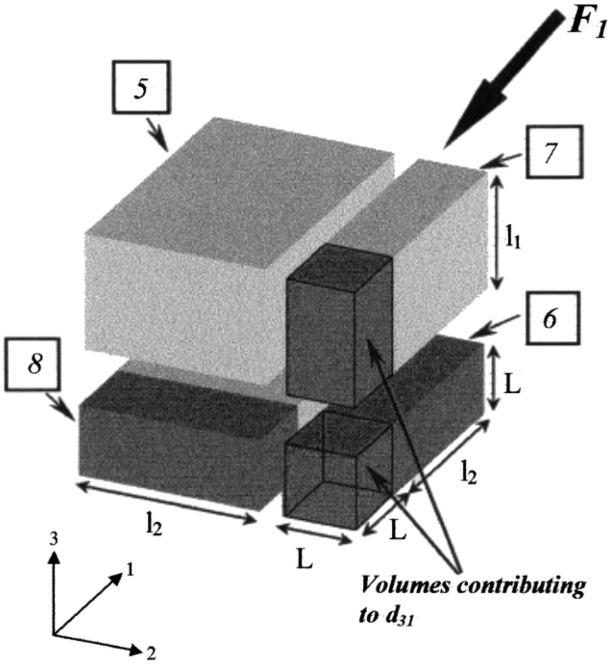

Fig. 2. Determination of d_{31}. As a force F_1 is applied, the load is distributed between volumes 5–8. The distribution depends of the respective cross-sectional areas and compliances.

The total volume of active material contributing to d_{31} of the composite is the cube in volume 6, as indicated in Fig. 2 (the rest of volume 6 is neglected as it is in series in the 3-direction with a polymer) and the piezoelectric phase in volume 7. Volume 5 does not contribute to d_{31} as it consists of only a polymer phase and the contribution of volume 8 is negligible as it is in series with a polymer phase in the 3-direction. The d_{31} of the unit composite ($d_{31}^{\mathrm{Composite}}$) is therefore given by:

$d_{31}^{\mathrm{Composite}} = d_{31}^{\mathrm{PZT}} \cdot \frac{\mathrm{L}}{(L+l_2)} \times$ fraction of force experienced by volumes 6 and 7

$$= d_{31}^{\mathrm{PZT}} \cdot \frac{L}{(L+l_2)} \cdot \left[\frac{a^{\mathrm{V6}}}{s_{11}^{\mathrm{V6}}} + \frac{a^{\mathrm{V7}}}{s_{11}^{\mathrm{V7}}}\right] \cdot \left[\frac{a^{\mathrm{V5}}}{s_{11}^{\mathrm{V5}}} + \frac{a^{\mathrm{V6}}}{s_{11}^{\mathrm{V6}}} + \frac{a^{\mathrm{V7}}}{s_{11}^{\mathrm{V7}}} + \frac{a^{\mathrm{V8}}}{s_{11}^{\mathrm{V8}}}\right]^{-1}$$
$$= d_{31}^{\mathrm{PZT}} \cdot \frac{L}{(L+l_2)} \cdot \left[\frac{L.l_1.(L+l_2)}{L.s_{11}^{\mathrm{PZT}} + l_2.s_{11}^{\mathrm{Polymer}}} + \frac{L^2}{s_{11}^{\mathrm{PZT}}}\right] \cdot$$
$$\left[\frac{l_1.l_2}{s_{11}^{\mathrm{Polymer}}} + \frac{L.(l_1+l_2).(L+l_2)}{L.s_{11}^{\mathrm{PZT}} + l_2.s_{11}^{\mathrm{Polymer}}} + \frac{L^2}{s_{11}^{\mathrm{PZT}}}\right]^{-1} \tag{7}$$

where $d_{31}^{\mathrm{PZT}}$is the d_{31} piezoelectric coefficient of the piezoelectric material. For examination of model outputs, the piezoelectric phase was PZT-5H, with $d_{33} = 593$ pC N^{-1}, $d_{31} = -274$ pC N^{-1}, $s_{33}^{\mathrm{PZT}} = 20.8$ pPa^{-1}, $s_{11}^{\mathrm{PZT}} = 16.4$ pPa^{-1} and $\varepsilon_{33}^{\mathrm{T}} = 3400\varepsilon_0$ where $\varepsilon_0 = 8.85\times10^{-12}$ F m^{-1}. The polymer was considered to be isotropic ($s_{11}^{\mathrm{Polymer}} = s_{33}^{\mathrm{Polymer}}$) and compliances in the range of 2.5×10^{-10} to 1×10^{-10} Pa^{-1} were examined.

3. Results

Fig. 3 shows the results of the variation of hydrostatic strain constant (d_{h}) versus polymer volume fraction. As the polymer volume fraction increases, d_{h} gradually increases to a maximum at around 50–60% polymer. The values of d_{h} have significantly improved when compared to monolithic value for PZT-5H, which is ~45 pC N^{-1} (as indicated in Fig. 3). It can be seen that increasing the compliance of the polymer phase leads to an increase in d_{h} for a given polymer volume fraction. This is due to the increased load experienced by the active piezoelectric phase as the elastic modulus of the polymer is reduced. A final observation from Figure 3 is that the maxima are fairly broad which aids in the production of the materials as very specific levels of polymer volume fraction are not required to produce high d_{h} composites.

The hydrostatic voltage constant (g_{h}) results are shown in Fig. 4. In this case there is a continuous increase in g_{h} as the polymer volume fraction is increased. In reality, as the

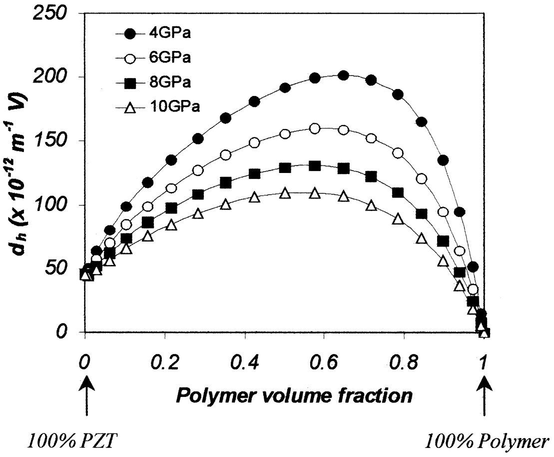

Fig. 3. Graph of hydrostatic strain constant (d_h) versus polymer volume fraction for a 3-3 piezocomposite. Results are shown for polymers of different elastic modulus.

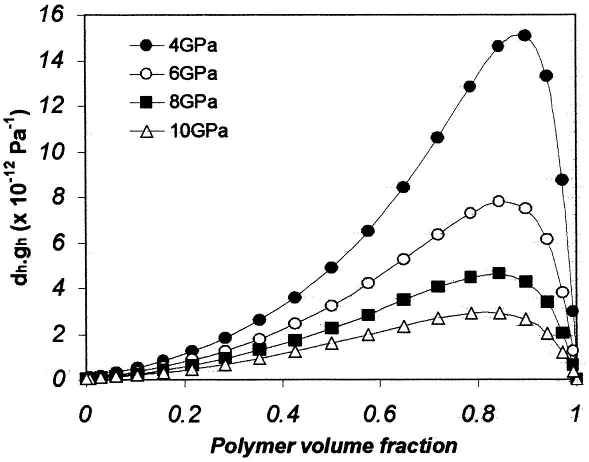

Fig. 4. Graph of hydrostatic voltage constant (g_h) versus polymer volume fraction for a 3-3 piezocomposite. Results are shown for polymers of different elastic modulus.

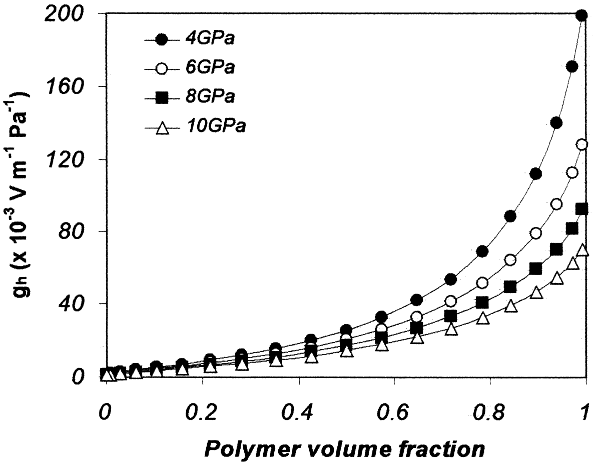

Fig. 5. Graph of hydrostatic figure of merit ($d_h \times g_h$) versus polymer volume fraction for a 3-3 piezocomposite. Results are shown for polymers of different elastic modulus.

polymer volume fraction increases to very high levels, the capacitance of the material is comparable to the electrical leads attached to the material and it is therefore unsuitable as a hydrophone or sensor.[5] This was shown to occur on 1–3 piezocomposites at polymer volume fraction greater than 96%.[5] In addition, as the polymer volume fraction is increased and the ceramic bears an increased load, depoling or mechanical damage of the piezoceramic phase becomes more likely. Again an increase in g_h is observed as the polymer compliance is increased due to improved stress transfer into the active ceramic phase.

The hydrostatic figure of merit (HFOM) results are shown in Fig. 5, calculated from the product $d_h \times g_h$. In this case there is a maximum in the HFOM at ~80% polymer volume fraction. The maxima are less broad than those observed in Fig. 5, requiring greater control and tolerances of polymer volume fraction if high $d_h \times g_h$ composites are to be manufactured. Again, larger values of the figure of merit are obtained by increasing the compliance of the passive phase.

4. Conclusions

An improved analytical model of a 3-3 piezocomposite structure has been presented. The model has shown that the necessary polymer volume fraction for optimised properties depends on the composite application and the figure of merit that needs to be maximised (d_h, g_h or $d_h \times g_h$). The results have shown that improvements in all figures of merit can be achieved by using a high compliance polymer phase due to increased stress transfer into the active piezoceramic phase. All of these observations agree well with reported values[7] and the model is a useful tool for examination of microstructural and materials properties on the behaviour of 3-3 piezocomposites. Pore anisotropy can be investigated by examination of various l_1/l_2 ratios and is the subject of further work. The limitations of the model are (i) the idealised structure of the model unit cell (which may not fully represent manufactured 3-3 piezocomposites), (ii) the assumption of the piezoelectric material being fully poled and (iii) the model considers the application of forces on each face separately, while under hydrostatic conditions the forces are applied simultaneously.

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