NONLINEAR ELECTROMECHANICAL RESPONSE AND POLARIZATION SWITCHING OF 1-3 PIEZOCOMPOSITES

Yasuhide Shindo, shindo@material.tohoku.ac.jp
Fumio Narita, narita@material.tohoku.ac.jp
Taiki Watanabe, a8tm5632@stu.material.tohoku.ac.jp
Department of Materials Processing, Graduate School of Engineering, Tohoku University
Aoba-yama 6-6-02, Sendai 980-8579, Japan

Abstract. This work deals with the nonlinear electromechanical response of 1-3 piezocomposites. The composites contain square or circular piezoelectric rods in an epoxy matrix. Experiments were performed to measure the displacement versus electric field curves. Three dimensional finite element analysis was also conducted to study the electromechanical fields in the 1-3 piezocomposites by introducing a model for polarization switching. Comparison was then made between numerical results and experimental data.

Keywords: Piezomechanics, Finite element method, Material testing, 1-3 piezoelectric/polymer composites, Electromechanical field concentrations

1. INTRODUCTION

Piezoelectric composites with 1-3 connectivity consist of piezoelectric rods or fibers in a polymer matrix. The advantages of the 1-3 piezocomposites are low acoustic impedance, low mechanical quality, and high electromechanical coupling coefficient. Therefore, they are well suited for ultrasonic transducers in non destructive testing and medical imaging applications. Recently, Sapsathiarn et al. (2008) analyzed the electro-mechanical interaction between a fiber and a matrix material in a 1-3 piezocomposite, and discussed the interfacial stresses for the representative composites. In some practical structures, one major concern has been the polarization switching of the piezoelectric rods or fibers. Also, theoretical and experimental investigations on the electromechanical response of the 1-3 piezocomposites are very limited.

In this work, we investigate theoretically and experimentally the nonlinear electromechanical response of 1-3 piezocomposites containing square or circular piezoelectric rods in an epoxy matrix. Nonlinear three dimensional finite element model incorporating the polarization switching mechanism is used to predict the electromechanical fields in the 1-3 piezocomposites. Displacement measurements are also recorded to characterize the response in the 1-3 piezocomposites. Results produced by the model are then compared with experimental values.

2. FINITE ELEMENT ANALYSIS

2.1. Basic equations

Consider a piezoelectric material with no body force and free charge. The governing equations in the Cartesian coordinates $x_i$ ($i = 1, 2, 3$) are

$$\sigma_{ji,j} = 0 \quad (1)$$
$$D_{i,i} = 0 \quad (2)$$

where $\sigma_{ji}$ is the stress tensor, $D_{i,i}$ is the electric displacement vector, a comma denotes partial differentiation with respect to the coordinate $x_i$, and the Einstein summation convention over repeated indices is used. The relation between the strain tensor $\varepsilon_{ij}$ and the displacement vector $u_i$ is given by

$$\varepsilon_{ij} = \frac{1}{2}(u_{j,i} + u_{i,j}) \quad (3)$$

and the electric field intensity vector is

$$E_i = -\phi_j \quad (4)$$
where \( \phi \) is the electric potential. In a ferroelectric material, domain switching leads to changes in the remanent strain \( \varepsilon_{ij}^r \) and remanent polarization \( P_i^r \). The constitutive relation can be written as

\[
\begin{align*}
\sigma_j &= c_{ijkl} (\varepsilon_{kl} - \varepsilon_{kl}^r) - e_{kl} E_k \\
D_j &= e_{ijkl} (\varepsilon_{kl} - \varepsilon_{kl}^r) + \varepsilon_{ik} E_k + P_i^r
\end{align*}
\]

In Eqs. (5) and (6), \( c_{ijkl} \) and \( e_{ijkl} \) are the elastic and piezoelectric tensors, and \( \varepsilon_{ik} \) is the dielectric permittivity tensor.

Valid symmetry conditions for the material constants are

\[
\begin{align*}
\varepsilon_{ik} &= \varepsilon_{kik}, \\
\varepsilon_{ij} &= \varepsilon_{jij}, \\
\varepsilon_{ik} &= \varepsilon_{kli}.
\end{align*}
\]

The constitutive equations (5) and (6) for piezoelectric material poled in the \( x_3 \)-direction are

\[
\begin{align*}
\begin{bmatrix}
\sigma_1 \\
\sigma_2 \\
\sigma_3 \\
\sigma_4 \\
\sigma_5 \\
\sigma_6
\end{bmatrix}
&= \begin{bmatrix}
\varepsilon_{11} & 0 & 0 & 0 & 0 & \varepsilon_{15} \\
0 & \varepsilon_{22} & 0 & 0 & 0 & 0 \\
0 & 0 & \varepsilon_{33} & 0 & 0 & 0 \\
0 & 0 & 0 & \varepsilon_{44} & 0 & 0 \\
0 & 0 & 0 & 0 & \varepsilon_{55} & 0 \\
0 & 0 & 0 & 0 & 0 & \varepsilon_{66}
\end{bmatrix}
\begin{bmatrix}
\varepsilon_{11} - \varepsilon_{11}^r \\
\varepsilon_{22} - \varepsilon_{22}^r \\
\varepsilon_{33} - \varepsilon_{33}^r \\
\varepsilon_{44} - \varepsilon_{44}^r \\
\varepsilon_{55} - \varepsilon_{55}^r \\
\varepsilon_{66} - \varepsilon_{66}^r
\end{bmatrix}
\begin{bmatrix}
0 & 0 & 0 \\
0 & 0 & \varepsilon_{15} \\
0 & 0 & 0 \\
0 & 0 & 0 \\
0 & 0 & 0 \\
0 & 0 & 0
\end{bmatrix}
\begin{bmatrix}
E_1 \\
E_2 \\
E_3
\end{bmatrix}
\end{align*}
\]

where

\[
\begin{align*}
\sigma_1 &= \sigma_{11}, \sigma_2 = \sigma_{22}, \sigma_3 = \sigma_{33}, \sigma_4 = \sigma_{44}, \sigma_5 = \sigma_{55}, \sigma_6 = \sigma_{66}, \sigma_{12} = \sigma_{21}, \\
\varepsilon_1 &= \varepsilon_{11}, \varepsilon_2 = \varepsilon_{22}, \varepsilon_3 = \varepsilon_{33}, \varepsilon_4 = 2\varepsilon_{23}, \varepsilon_5 = 2\varepsilon_{31}, \varepsilon_6 = 2\varepsilon_{13}, \\
c_{11} &= c_{1111}, c_{12} = c_{1122}, c_{13} = c_{1113}, c_{22} = c_{2222}, c_{23} = c_{2233}, c_{33} = c_{3333}, c_{44} = c_{2323} = c_{3131}, \\
c_{66} &= \frac{1}{2} (c_{11} - c_{22}) \\
e_{15} &= e_{131} = e_{223}, e_{31} = e_{311}, e_{32} = e_{322}, e_{33} = e_{333}
\end{align*}
\]

The enhanced electromechanical field level results in localized polarization switching. The criterion states that a polarization switches when the electrical and mechanical work exceeds a critical value (Hwang et al., 1995)

\[
\sigma_j \Delta \varepsilon_{ij} + E_j \Delta P_i^r \geq 2P^r E_c
\]
where $P_s$ is a spontaneous polarization, $E_c$ is a coercive electric field, and $\Delta \epsilon_{ij}$ and $\Delta P_i$ are the changes in the spontaneous strain and spontaneous polarization during switching, respectively. The values of $\Delta \epsilon_{ij} = \epsilon'_{ij}$ and $\Delta P_i = P'_i$ for $180^\circ$ switching can be expressed as

$$
\begin{align*}
\Delta \epsilon_{11} &= 0, & \Delta \epsilon_{22} &= 0, & \Delta \epsilon_{33} &= 0, & \Delta \epsilon_{12} &= 0, & \Delta \epsilon_{23} &= 0, & \Delta \epsilon_{31} &= 0, \\
\Delta P_1 &= 0, & \Delta P_2 &= 0, & \Delta P_3 &= -2P'
\end{align*}
$$

(12)

For $90^\circ$ switching in the $x_3x_1$ plane, the changes are

$$
\begin{align*}
\Delta \epsilon_{11} &= \gamma', & \Delta \epsilon_{22} &= 0, & \Delta \epsilon_{33} &= -\gamma', & \Delta \epsilon_{12} &= 0, & \Delta \epsilon_{23} &= 0, & \Delta \epsilon_{31} &= 0, \\
\Delta P_1 &= \pm P', & \Delta P_2 &= 0, & \Delta P_3 &= -P'
\end{align*}
$$

(13)

where $\gamma'$ is a spontaneous strain. For $90^\circ$ switching in the $x_3x_3$ plane, we have

$$
\begin{align*}
\Delta \epsilon_{11} &= 0, & \Delta \epsilon_{22} &= \gamma', & \Delta \epsilon_{33} &= -\gamma', & \Delta \epsilon_{12} &= 0, & \Delta \epsilon_{23} &= 0, & \Delta \epsilon_{31} &= 0, \\
\Delta P_1 &= \pm P', & \Delta P_2 &= 0, & \Delta P_3 &= -P'
\end{align*}
$$

(14)

The constitutive equations (5) and (6) after polarization switching are given by

$$
\begin{align*}
\sigma_{ij} &= e_{ijkl} (\epsilon_{kl} - \epsilon'_k) - e'_{ijkl} E_k \\
D_j &= e'_{ijkl} (\epsilon_{kl} - \epsilon'_k) + \epsilon_{ikl} E_k + P'_i
\end{align*}
$$

(15)

(16)

The new piezoelectric constant $e'_{ijkl}$ is related to the elastic and direct piezoelectric constants by

$$
\begin{align*}
e'_{11} &= d'_{111} c_{11} + d'_{122} c_{12} + d'_{133} c_{13}, & e'_{12} &= d'_{111} c_{12} + d'_{122} c_{12} + d'_{133} c_{13}, \\
e'_{13} &= d'_{111} c_{13} + d'_{122} c_{13} + d'_{133} c_{33}, & e'_{14} &= 2d'_{123} c_{44}, & e'_{15} &= 2d'_{131} c_{44}, & e'_{16} &= 2d'_{112} c_{66}, \\
e'_{21} &= d'_{211} c_{11} + d'_{222} c_{12} + d'_{233} c_{13}, & e'_{22} &= d'_{211} c_{12} + d'_{222} c_{11} + d'_{233} c_{13}, \\
e'_{23} &= d'_{211} c_{13} + d'_{222} c_{13} + d'_{233} c_{33}, & e'_{24} &= 2d'_{223} c_{44}, & e'_{25} &= 2d'_{231} c_{44}, & e'_{26} &= 2d'_{212} c_{66}, \\
e'_{31} &= d'_{311} c_{11} + d'_{322} c_{12} + d'_{333} c_{13}, & e'_{32} &= d'_{311} c_{12} + d'_{322} c_{11} + d'_{333} c_{13}, \\
e'_{33} &= d'_{311} c_{13} + d'_{322} c_{13} + d'_{333} c_{33}, & e'_{34} &= 2d'_{323} c_{44}, & e'_{35} &= 2d'_{331} c_{44}, & e'_{36} &= 2d'_{312} c_{66}
\end{align*}
$$

(17)

The components of the piezoelectricity tensor $d'_{ijkl}$ are

$$
\begin{align*}
d'_{ijkl} &= \left\{ d_{33} n_i n_j n_k + d_{31} (n_i \delta_{jk} - n_j n_k n_l) + \frac{1}{2} d_{15} (\delta_{jk} n_i - 2n_j n_k n_l + \delta_{il} n_k) \right\}
\end{align*}
$$

(18)

where $n_i$ is the unit vector in the poling direction, $\delta_{ij}$ is the Kroneker delta, and $d_{33} = d_{333}$, $d_{31} = d_{311}$, $d_{15} = 2d_{131}$ are the direct piezoelectric constants.

2.2. Computational model

A three-dimensional finite element analysis (ANSYS) was performed to represent the electromechanical field distributions of the 1-3 piezocomposites. Consider two classes of 1-3 piezocomposites (see Fig.1). A rectangular
Cartesian coordinate system \((x, y, z)\) is used with the \(z\)-axis coinciding with the poling direction. \(4N^2\) square piezoelectric rods of side \(a\) and height \(h\) (Fig. 1 (a)) or circular piezoelectric rods of diameter \(d\) and height \(h\) (Fig. 1 (b)) are embedded in a polymer matrix, and arranged in a square array; where \(N\) is half of the number of rods aligned in the \(x\)- or \(y\)-direction. The height of the 1-3 piezocomposites is \(h\). Repeating units of the composites with \(4N^2\) square and circular piezoelectric rods occupy the region \((0 \leq x \leq a + ag, 0 \leq y \leq a + ag, 0 \leq z \leq h/2)\) and \((0 \leq x \leq d + dg, 0 \leq y \leq d + dg, 0 \leq z \leq h/2)\) and the volume fractions \(V_f\) of the square- and circular-type 1-3 piezocomposites are \(a^2/(a + ag)^2\) and \(\pi d^2/4(d + dg)^2\), respectively, where \(ag\) and \(dg\) are the gaps between the piezoelectric rods. Electrodes lie in the top and bottom ends of each piezoelectric rod. The electric potential on the top electrode surfaces equals the applied voltage, \(\phi = V_0\). The bottom electrode surfaces are connected to the ground, so that \(\phi = 0\). Because of the geometric and loading symmetry, only one-eighth of the specimen needs to be analyzed. The switching criterion of Eq. (11) is checked for every element and for every possible polarization direction to see if switching will occur. After all possible polarization switches have occurred, the piezoelectricity tensor of each element is rotated to the new polarization direction (Shindo et al., 2004, Narita et al., 2005). The spontaneous polarization \(P_s\) and strain \(\gamma_s\) are assigned representative values of 0.3 C/m² and 0.004, respectively.

![Figure 1. Illustration of 1-3 piezocomposite with (a) square and (b) circular rods](image)

### 3. EXPERIMENTAL PROCEDURE

The 1-3 piezocomposites \((N = 2)\) were fabricated using 16 square soft lead zirconate titanate (PZT) rods C-6 and epoxy resin P80B10. The material properties of C-6 are listed in Table 1 (Fuji Ceramics Co., Ltd., Japan), and the coercive electric field is approximately \(E_c = 0.45\) MV/m. Young’s modulus and Poisson’s ratio of P80B10 are 650 MPa and 0.35, respectively. Electrodes are attached on the top and bottom surfaces of the composite to address each rod. The dimension of the specimen is 32 mm × 32 mm × 27 mm. The piezoelectric rods have side \(a = 7\) mm and height \(h = 27\) mm. The gap is \(ag = 1\) mm. So the volume fraction \(V_f\) is about 0.77. The specimen was placed on the rigid floor. Voltage was applied using a power supply, and a digital microscope at ×1000 magnification was used to measure the magnitude of the deformation.

<table>
<thead>
<tr>
<th>Elastastic stiffnesses ((\times 10^9\text{N/m}^2))</th>
<th>Piezoelectric coefficients ((\text{C/m}^2))</th>
<th>Dielectric constants ((\times 10^{-11}\text{C/Vm}))</th>
</tr>
</thead>
<tbody>
<tr>
<td>(c_{11})</td>
<td>(c_{33})</td>
<td>(c_{44})</td>
</tr>
<tr>
<td>C-6</td>
<td>12.3</td>
<td>11.2</td>
</tr>
</tbody>
</table>
4. RESULTS AND DISCUSSION

We present results for the 1-3 piezocomposites with square or circular PZT C-6 rods. The height of the composites is held constant \( (h = 27 \text{ mm}) \). Fig.2 shows the displacement \( u_z \) versus electric field \( E_0 = -V_0/h \) for the square-type 1-3 piezocomposite of \( a = 7 \text{ mm}, \ a_g = 1 \text{ mm} \ (V_f \sim 0.77) \) and \( N = 2 \) at the measured point \((x = 16 \text{ mm}, \ y = 4 \text{ mm}, \ z = 13.5 \text{ mm})\). The solid line represents the finite element analysis (FEA) result while the solid circle represents the experimental data. Also shown is the FEA result for \( N = \infty \) (dashed line). The number of piezoelectric rods has little effect on the displacement. As the electric field \( E_0 \) is reduced from zero initially, the negative displacement increases. It can be seen that an applied electric field of approximately \(-0.445 \text{ MV/m}\) causes polarization switching in the portion of the specimen. The polarization switching can cause an unexpected decrease in the negative displacement and the sudden increase of positive displacement during switching. As the magnitude of positive electric field increases, the positive displacement increases. The trend is similar between the analysis and experiment. Fig.3 shows the displacement \( u_z \) versus electric field \( E_0 \) for the circular-type 1-3 piezocomposite of \( d = 7.95 \text{ mm}, \ d_g = 0.05 \text{ mm} \ (V_f \sim 0.77) \) and \( N = 2 \) at \( x = 0 \text{ mm}, \ y = 0 \text{ mm}, \ z = 13.5 \text{ mm} \), obtained from the FEA. Similar to the case of square-type 1-3 piezocomposite, nonlinear behavior is observed under negative electric fields. Fig.4 shows the polarization switching zones at the top surface \((z = 13.5 \text{ mm})\) of the (a) square- and (b) circular-type 1-3 piezocomposite under \( E_0 = -0.446, -0.45 \text{ MV/m} \). The size of the 180° switching zone increases with increasing electric field opposite to the original poling direction. The 180° switching is complete at electric field strength of about \(-0.470 \text{ MV/m} \) (not shown). The 90° switching does not occur.

![Figure 2. Displacement versus electric field for square-type 1-3 piezocomposite](image)

![Figure 3. Displacement versus electric field for circular-type 1-3 piezocomposite](image)
In conclusion, the electric field induced displacement showed nonlinear behavior because of localized polarization switching. Also, the displacement for the 1-3 piezocomposites containing square piezoelectric rods was large compared to the composites with circular piezoelectric rods.

REFERENCES


RESPONSIBILITY NOTICE

The authors are the only responsible for the printed material included in this paper.