Challenges and New Trends for Piezoelectric Actuators

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Polycrystalline lead zirconate titanate (PZT) has been the material of choice for piezoelectric applications such as medical ultrasound, sonar, fuel modulation, and active vibration damping; however, there have been no significant improvements in properties in recent years in this now mature material system. The main focus in the piezoelectric material development beyond the capabilities of PZT has been two-fold: (i) increasing the electromechanical activity, and (ii) increasing the operating temperature.

(i) Remarkable properties have been reported in recent years for an emerging class of piezoelectric crystals based on lead magnesium niobate (PMN) substituted with lead titanate (PT). These new material systems can have exceptionally large piezoelectric coefficients and electromechanical coupling factors (e.g., $k_{33}=0.94$). The latter implies over 90% electro-mechanical energy conversion ($k^2$), and the former are approximately an order-of magnitude greater than commercially available piezoelectrics based on polycrystalline PZT. Recently, developments in the growth of PMN-PT single crystals and discovery of a unique direction among the crystallographically equivalent axes have made it feasible to use single crystal elements in high-end piezoelectric applications.

(ii) Development of high temperature piezoceramics has been a tougher challenge to meet. Most high temperature piezoelectric material systems suffer from low piezoelectric coefficients and are only suitable for sensor applications. Two main challenges in producing high temperature piezoelectrics are (a) to increase Curie temperature ($T_C$) without an increase in loss tangent, and (b) to demonstrate high piezoelectric activity. BiScO$_3$-PbTiO$_3$ (BS-PT) based systems exhibited high $T_C$ and large piezoelectric coefficients near the morphotropic phase boundary (MPB). The effects of excess PbO and Bi$_2$O$_3$ and their partitioning in grain boundaries were studied using impedance spectroscopy, ferroelectric, and piezoelectric measurement techniques. Excess Bi$_2$O$_3$ proved to be a successful liquid phase forming additive to improve the BS-PT piezoceramics for high temperature applications, as a result of increased resistivity and enhanced piezoelectric activity through microstructure engineering.
Challenges and New Trends for Piezoelectric Actuators

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Outline

Challenges and new materials
High piezoelectric coefficient
Unique direction
High operating temperature

H.C. Materials Corp.
COMPARISON:
• Same muscle tissue powers both insects and elephants. But, muscle but does not work at high temperatures and frequencies.

• Gas turbine applications require high frequency and displacement at elevated temperatures.

What does Piezoelectric offer?

Poling

All materials (32)

Piezoelectrics (20)
Non-centrosymmetric

Pyroelectrics (10)
Polar

Ferroelectrics
Reorientable domains
PMN-PT

- Remarkable properties in the MPB region.

Crystal numbers

1 \rightarrow 6

Critical point (CP)

MPB

Dielectric properties of $<001>$ poled crystals

<table>
<thead>
<tr>
<th>Specimen</th>
<th>$K_{33}$</th>
<th>tan $\delta$</th>
<th>$d_{33}$ (pC/N)</th>
<th>$k_{33}$</th>
<th>$R \rightarrow T$</th>
<th>$T \rightarrow C$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>5000</td>
<td>0.002</td>
<td>1200</td>
<td>0.914</td>
<td>104</td>
<td>112</td>
</tr>
<tr>
<td>2</td>
<td>5625</td>
<td>0.002</td>
<td>1475</td>
<td>0.920</td>
<td>105</td>
<td>113</td>
</tr>
<tr>
<td>3</td>
<td>6025</td>
<td>0.003</td>
<td>1625</td>
<td>0.927</td>
<td>105</td>
<td>120</td>
</tr>
<tr>
<td>4</td>
<td>8465</td>
<td>0.002</td>
<td>3470</td>
<td>0.934</td>
<td>97</td>
<td>129</td>
</tr>
<tr>
<td>5</td>
<td>9195</td>
<td>0.004</td>
<td>3620</td>
<td>0.939</td>
<td>97</td>
<td>132</td>
</tr>
<tr>
<td>6</td>
<td>14260</td>
<td>0.005</td>
<td>4080</td>
<td>0.951</td>
<td>87</td>
<td>137</td>
</tr>
<tr>
<td>PZT 5H</td>
<td>3400</td>
<td>0.025</td>
<td>590</td>
<td>0.75</td>
<td>195</td>
<td></td>
</tr>
</tbody>
</table>

Outside the MPB region, <001> direction

Unpoled

In the MPB region, <001> direction

Diffuse transformation is not due to relaxor behavior
Effect of poling, $<001>$ direction

**Close to the CP**

**Close to the MPB**

Effect of poling on phase transformation temperatures

Poling along $<001>$
Summary (Dielectric behavior)

• Diffuse $K_{33}$ vs. $T$ transformation without frequency dispersion
  (convergence of $R \rightarrow T \rightarrow C$ phases)

• Effects of poling along $<001>$
  – increases $K_{33}$ at room temperature
  – decreases the $R \rightarrow T$ transformation temperature
  – increases $K_{33}$ at $R \rightarrow T$

• Poling does not have any effect on the dielectric behavior of the $T \rightarrow C$ phase transformation.

Domain orientations, unpoled

- $\alpha = \beta = \gamma \neq 90$
  - Rhombohedral
- $\alpha = \beta = \gamma = 90$
  - Tetragonal
- $\alpha = \beta = \gamma = 90$
  - Cubic
Effect of orientation, <001>, unpoled

All along <001>

<table>
<thead>
<tr>
<th></th>
<th>Along 1 (ppm/K)</th>
<th>Along 2 and 3 (ppm/K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\alpha^R$</td>
<td>-8.6</td>
<td>4.5</td>
</tr>
<tr>
<td>$\alpha^T$</td>
<td>-25</td>
<td>14</td>
</tr>
<tr>
<td>$\alpha^C$</td>
<td>17</td>
<td>17</td>
</tr>
<tr>
<td>$K_{33}$</td>
<td>5424</td>
<td>1765</td>
</tr>
</tbody>
</table>

$2^\circ$C/min, heating

Unpoled


Effect of poling – measured parallel to poling

Unpoled

Poled

[001]
Effect of poling - measured perpendicular to poling direction

Effect of poling (near critical point)

- Poling reorients the unique direction
- $\alpha$ shows uniaxial macroscopic symmetry
- Depoled is the same as unpoled

$K_{33} = 8430$ (poled parallel)

$K_{33} = 4573$ (poled normal)
Effect of composition (unpoled crystal)

Close to the MPB

<table>
<thead>
<tr>
<th>$K_{33}$</th>
<th>Unique</th>
<th>Normal 1</th>
<th>Normal 2</th>
</tr>
</thead>
<tbody>
<tr>
<td>MPB</td>
<td>5425</td>
<td>1955</td>
<td>1765</td>
</tr>
<tr>
<td>Critical point</td>
<td>4870</td>
<td>4710</td>
<td>4685</td>
</tr>
</tbody>
</table>

Close to the critical point

Effect of poling on T$\rightarrow$C temperature

Open circuit

$\Delta T = 30^\circ$C
**Boundary conditions**

*Open circuit* \((\Delta P=0)\)

*Short circuit* \((\Delta E=0)\)

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**T\(\rightarrow\)C phase transformation**

- **Unpoled**
  - 141\(^\circ\)C
  - 142\(^\circ\)C

- **Poled**
  - 142\(^\circ\)C
  - 150\(^\circ\)C

Scale: 0.5 mm
Summary

• not all of the <001> directions were macroscopically equivalent even though they were indistinguishable by XRD.
• the anomaly may be attributable to the method of seeded crystal growth.
• The unique direction was related to the domain structure of the crystal and could be reoriented by room temperature poling in a direction normal to the unique direction.
• Highest K is obtained when poled along the unique direction
• Poling promoted the formation of c-oriented tetragonal domains along the poling direction.
• the phase transformations were sharper under short-circuit boundary conditions.

Objective

Development of high-temperature piezoelectric actuators for aeronautics and aerospace applications.

Applications

• Actuators for Aerospace and Aeronautics
  – Fuel modulation, valves, micro-positioning devices, MEMS, active damping and energy harvesting.
• Sensors
  – Pressure sensors, passive damping
Challenges for High Temperature

- Trade off between $T_C$ and $d_{33}$
- Conductivity at elevated temperatures

<table>
<thead>
<tr>
<th>Material</th>
<th>$T_C$ (°C)/(°F)</th>
<th>$d_{33}$ (pC/N)</th>
</tr>
</thead>
<tbody>
<tr>
<td>PZT Type II (PZT 5A)</td>
<td>350 / 662</td>
<td>374</td>
</tr>
<tr>
<td>PMN-PT single crystals</td>
<td>90 / 194</td>
<td>&gt;2000</td>
</tr>
<tr>
<td>BiScO$_3$-PbTiO$_3$</td>
<td>450 / 842</td>
<td>401</td>
</tr>
<tr>
<td>La$<em>3$Ga$</em>{15.5}$Ta$<em>{0.5}$O$</em>{14}$ single crystal</td>
<td>N/A</td>
<td>7</td>
</tr>
<tr>
<td>Na$<em>{0.5}$Bi$</em>{14.5}$TiO$_5$</td>
<td>650 / 1202</td>
<td>19</td>
</tr>
<tr>
<td>La$_3$Ti$_4$O$_7$</td>
<td>1482 / 2700</td>
<td>16</td>
</tr>
</tbody>
</table>


Processing of BS-PT

1. Raw materials (Bi$_2$O$_3$, PbO, Sc$_2$O$_3$, TiO$_2$)
2. Ball milling (15hrs)
3. Drying (stirred)
4. Calcination (750°C, 3hrs, 5°C/min), in air
5. Ball milling (6hrs)
6. Excess addition
7. Pressing
8. Sintering (1100°C, 1hr, 5°C/min), in air
Sintering Conditions for BS-PT

- 615 °C
- 1100 °C
- 1200 °C
- 1280 °C

Netzch horizontal dilatometer
5°C/min, heating, air, alumina push rod

Electrical characterization

- Impedance measurements *(Solartron and HP Agilent)*
  - 1Hz-1MHz, Room temperature to 1000 °C.
  - 40Hz-110MHz, Room temperature to 600 °C.
  - 1Mhz-3Ghz, Microwave range
    (Determination of electrical, dielectric and electromechanical properties)

- Ferroelectric measurements *(Radiant Technologies)*
  - Bipolar, unipolar loops, leakage (up to 10,000V)

- Piezoelectric measurements
  - Laser dopplermeter *(Polytech)* coupled with a signal generator and a high power amplifier (up to 10,000V)
  - PhotonicTM sensor *(MTI technologies)* coupled with Radiant
  - Berlincourt d$_{33}$ -meter
**Impedance Spectroscopy**  
*Conductivity - Grain/Grain Boundary*

\[ R \parallel C : \text{Resonance frequency } \omega_0 = \frac{1}{RC} \]

- Conduct: grain ≠ grain boundary  
  - \[ \omega_0 \text{ grain boundary} << \omega_0 \text{ grain-Low T} \]
  - \[ \omega_0 \text{ grain boundary} \sim \omega_0 \text{ grain-High T} \]

**Effect of Pb on microstructure**

0% Pb  
2% Pb  
5% Pb
Effect of Pb in BS-PT

Grain boundary contribution

10 kHz, 0.5 V/mm ac, in air

<table>
<thead>
<tr>
<th>Relaxation frequency</th>
<th>0% Pb</th>
<th>2% Pb</th>
<th>5% Pb</th>
</tr>
</thead>
<tbody>
<tr>
<td>800 Hz</td>
<td>3 kHz</td>
<td>5 kHz</td>
<td></td>
</tr>
</tbody>
</table>

Effect of Pb in BS-PT

<table>
<thead>
<tr>
<th></th>
<th>$\rho_{\text{grain}}$ ($\Omega\cdot\text{cm}$)</th>
<th>Dielectric constant, $K_{\text{grain}}$</th>
<th>$E_{\text{ac}}$ (ev)</th>
</tr>
</thead>
<tbody>
<tr>
<td>No excess</td>
<td>$4.1 \times 10^{10}$</td>
<td>1085</td>
<td>0.36</td>
</tr>
<tr>
<td>2% Pb</td>
<td>$2.4 \times 10^{10}$</td>
<td>805</td>
<td>0.35</td>
</tr>
<tr>
<td>5% Pb</td>
<td>$1.4 \times 10^{10}$</td>
<td>727</td>
<td>0.35</td>
</tr>
</tbody>
</table>
Effect of Bi on microstructure

0% Bi

2% Bi

5% Bi

10% Bi

Effects of Bi in BS-PT

1 kHz, 0.5 V/mm ac, in air
### Effect of Bi in BS-PT

<table>
<thead>
<tr>
<th></th>
<th>$\rho_{\text{grain}}$ (Ω.cm)</th>
<th>Dielectric constant, $K_{\text{grain}}$</th>
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<tr>
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<td>$4.1 \times 10^{10}$</td>
<td>1085</td>
<td>800 Hz</td>
<td>0.36</td>
</tr>
<tr>
<td>2% Bi</td>
<td>$4.9 \times 10^{10}$</td>
<td>1104</td>
<td>400 Hz</td>
<td>0.40</td>
</tr>
<tr>
<td>5% Bi</td>
<td>$19.6 \times 10^{10}$</td>
<td>1402</td>
<td>200 Hz</td>
<td>0.38</td>
</tr>
</tbody>
</table>

### Ferroelectric and piezoelectric properties

**Polarization (µC/cm²)**

- No excess
- 5% Bi
- 5% Pb

**E-field (kV/cm)**

- 100 °C
Ferroelectric Properties

No excess

$E_C = 13.5 \text{ kV/cm}$

$100\, ^\circ \text{C}$

5% Bi excess

Unipolar frequency dependence

5% Bi excess

$100\, ^\circ \text{C}$

No excess

$100\, ^\circ \text{C}$
Piezoelectric coefficient

Strain (%)

- 5% Bi excess
- No excess

\[ d_{33} = 408 \text{ pC/N} \]
\[ d_{33} = 354 \text{ pC/N} \]

High field resistivity

Resistivity (\(\Omega\text{.cm}\))

- No excess
- 5% Bi

\[ 100 \degree C \]
\[ 0 \rightarrow 50 \text{ kV/cm} \]
Summary

• BiScO$_3$-PbTiO$_3$ ceramics with $T_C > 400^\circ$C has been successfully processed.
• Despite the increase in $T_C$, excess Pb addition increases both the bulk conductivity and the grain boundary contribution to conductivity at elevated temperatures.
• Conductivity at elevated temperatures, that limits the operating temperature for actuators, has been greatly reduced by excess Bi additions.
• Excess Bi doping improves poling conditions resulting in enhanced piezoelectric coefficient ($d_{33} = 408$ pC/N).

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