Piezoelectric Material to Perform Ultrasonic Non-Destructive Evaluation within an Operating Reactor Core.

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General Requirements

- High temperature capability
  - Hard ferroelectrics
    - Bismuth Titanate
    - Lithium Niobate
      - Relative Permittivity < 30, \( \rho \ 10^6 \text{ – } 10^8 \ \Omega \text{cm} \)
  - Polar Single Crystals
    - AlN
    - ZnO
    - GaPO_4
      - Relative Permittivity < 30, \( \rho \ 10^3 \text{ – } 10^9 \ \Omega \text{cm} \)
- Low relative permittivity and low resistivity can be detrimental
- Radiation Resistant
- Easy disposal
## Literature Review

<table>
<thead>
<tr>
<th>Material</th>
<th>Fast Neutron Fluence [n/cm^2]</th>
<th>Thermal Fluence [n/cm^2]</th>
<th>k</th>
<th>Conversion</th>
<th>tan(δ)</th>
<th>C</th>
<th>R</th>
<th>Temp [°C]</th>
</tr>
</thead>
<tbody>
<tr>
<td><em>PbZr_{0.51} Ti_{0.463} Nb_{0.02} Li_{0.007} O_3 (soft)</em></td>
<td>10^{18}</td>
<td>-30%</td>
<td>-45%</td>
<td>-75%</td>
<td>70</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td><em>PbZr_{0.45} Ti_{0.49} Mn_{0.17} Nb_{0.033} O_3 (hard)</em></td>
<td>10^{18}</td>
<td>-25%</td>
<td>-45%</td>
<td>-75%</td>
<td>70</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>Bi_{3}TiNbO_{9} +0.3% Cr_{2}O_{3}</strong></td>
<td>1.5X10^{20}</td>
<td>3X10^{20}</td>
<td>-60%</td>
<td>-10%</td>
<td>-50%</td>
<td>2 to 72</td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>TsTs-21 (aka PZT)</strong></td>
<td>1.5X10^{20}</td>
<td>3X10^{20}</td>
<td>-75%</td>
<td>-50%</td>
<td>-70%</td>
<td>2 to 72</td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>TSVM-1</strong></td>
<td>1.9X10^{18}</td>
<td>1.8X10^{19}</td>
<td>-10%</td>
<td>100%</td>
<td>-20%</td>
<td>2 to 72</td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>LiNbO_3</strong></td>
<td>1.85X10^{18}</td>
<td>5.8X10^{18}</td>
<td>-75%</td>
<td>0</td>
<td>15%</td>
<td>-8%</td>
<td>8%</td>
<td>25 to 225</td>
</tr>
</tbody>
</table>

- * C. Miclea, et. al. 2005
- **Meleshko, et al. 1980s**
- ***Primak, et al 1976***
Radiation Damage

• Elastic/Inelastic scattering
  • displaced atoms
  • heating

• Transmutation reaction
  • displaced atoms
  • heating
  • foreign atoms (hydrogen, helium and daughter nucleus)
Heuristic Overview of AlN Radiation Hardness

• No phase transition aside from melt at 2800° C
  • Thermal spiked regions relax/anneal to piezoelectric state
    • AlN has been produced in this state (Wurtzite) via 190 keV N⁺ ion irradiation of Al

• Cross section for charged particle reactions is small

• dpa due to fast neutrons is very nearly stoichiometric
Displaced Atoms (Elastic Scattering)
(Kinchin Pease model + ENDF data)

<table>
<thead>
<tr>
<th>Material</th>
<th>Atom</th>
<th>dpa/10^{22} n/cm²</th>
<th>Total dpa/10^{22} n/cm²</th>
</tr>
</thead>
<tbody>
<tr>
<td>AlN</td>
<td>Al</td>
<td>1.99</td>
<td>4</td>
</tr>
<tr>
<td></td>
<td>N</td>
<td>1.97</td>
<td></td>
</tr>
<tr>
<td>LiNbO₃</td>
<td>Li</td>
<td>1.47</td>
<td>3.7</td>
</tr>
<tr>
<td></td>
<td>Nb</td>
<td>0.47</td>
<td></td>
</tr>
<tr>
<td></td>
<td>O</td>
<td>1.79</td>
<td></td>
</tr>
<tr>
<td>Bi₄Ti₃O₁₂</td>
<td>Bi</td>
<td>0.71</td>
<td>3.4</td>
</tr>
<tr>
<td></td>
<td>Ti</td>
<td>0.58</td>
<td></td>
</tr>
<tr>
<td></td>
<td>O</td>
<td>2.1</td>
<td></td>
</tr>
<tr>
<td>PZT 5A</td>
<td>Pb</td>
<td>1.15</td>
<td>3.25</td>
</tr>
<tr>
<td></td>
<td>Zr</td>
<td>0.66</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Ti</td>
<td>0.55</td>
<td></td>
</tr>
<tr>
<td></td>
<td>O</td>
<td>0.89</td>
<td></td>
</tr>
</tbody>
</table>
Transmutation Reactions

<table>
<thead>
<tr>
<th>Material</th>
<th>LiNbO₃</th>
<th>AlN</th>
<th>Bi₄Ti₃O₁₂</th>
</tr>
</thead>
<tbody>
<tr>
<td>(n,α)</td>
<td>14.9</td>
<td>N/A</td>
<td>&lt;0.01</td>
</tr>
<tr>
<td>(n,p)</td>
<td>&lt;0.01</td>
<td>0.07</td>
<td>N/A</td>
</tr>
</tbody>
</table>

- **Macroscopic cross section**
  - $\mu = \sum_i N_i \sigma_{ij}$

- $N_i$ atomic density including all naturally occurring isotopes ($^6$Li at %7.5)

- $j$ represents reaction channel in this case total proton or alpha particle production (MT 203 and 207)

Determined as the cause of degradation in LiNbO₃ (Primak 1976)

Maxwellian spectrum used to calculate spectral averaged cross section (centered at thermal energy 0.025 eV)

Isotope tailoring?
Experimental Setup

- Radiation hardened cable
- Alumina insulation
- Al screw on cap
- Al cylinder
- Carbon backing
- Al plunger/electrical contact
- AlN
Basic Results of Radiation Hardness of AlN

- Pulse Echo Amplitude with Reactor Turned off

- Only change is a minor increase
- Amplitude holds up well
- Data collected over 3 months
Basic Results of Radiation Hardness

- Waveforms with reactor turned off before and after fluence

- A defected cable was replaced prior to top right waveform acquisition

- Aside from increase in amplitude due to cable replacement amplitude remains virtually unchanged

- The ringing is the primary change observed
Basic Results of Radiation Hardness

- Decrease in bandwidth is apparent for high fluence
- Possible Causes
  - Deterioration of backing material
  - Deterioration of coupling to aluminum cylinder
  - Damage to crystal
Transient Effects

- Amplitude drops during irradiation
- Dielectric loss?
- Reduced coupling between piezo and Al cylinder (loss of contact)?
Transient Effects

- Dielectric loss changes during irradiation by a negligible amount

- Both Q and dielectric loss are consistent with reduced coupling to Al cylinder

- Should have used a spring to reduce this effect (due to heating)
Measuring Temperature via Time of Flight

- Threshold crossing utilized to measure arrival time
- Secondary echo on transit 2 makes peak methods inapplicable as Q increases
Gamma Heating Measurement

- Theory - Newton’s law of cooling with and without constant source

- $T - T_{\text{ambient}} = \frac{q}{k} (1 - e^{-kt})$
- $T - T_{\text{ambient}} = (T_{\text{initial}} - T_{\text{ambient}}) e^{-kt}$
Second Harmonic in Al Cylinder

- Preliminary data
- Accounted for the increases in $Q$
- Heat treatment or atomic displacement may be the cause
Conclusion

• AlN is uneffected by a fast and thermal neutron fluence of $1.85 \times 10^{18}$ n/cm$^2$ and $5.8 \times 10^{18}$ n/cm$^2$ respectively and a gamma dose of 26.8 MGy
  • Also incidentally cycling to 200$^\circ$ C nearly 200 times

• Post irradiation testing is simplified due to lack of activity >1μRad/hr at 10 cm

• Post irradiation testing revealed $d_{33}$ as 5.5 pC/N (upper end of literature values for pristine samples)

• Not only does the transducer survive but it makes measurements in the process

• Taking the fluence up higher may be more convincing (our fast fluence was “very fast”)
AlN at High Temperature

• Performance as a high temperature MHz ultrasonic transducer varies

• It is believed that conduction due to defects/impurities is to blame

• Conductivity of AlN single crystals varies
  • Typically used in LED and semiconductor applications
  • Large band gap is the primary attraction

• $d_{33}$ varies as well
  • our samples 1-6 pC/N
AlN at High Temperature

Resistivity and high temperature transduction show a strong correlation

Crystal A

Crystal B

Resistivity $\Omega \cdot m$

Temperature $C^\circ$ x $10^3$

Peak to Peak Amplitude [V]

Temperature $C^\circ$

not sure on units
Piezoelectric Screening

- Resistivity and capacitance are equally important
- RC constant or tanδ increase dramatically with temperature in poor crystals

Screening Effect

- \( k^{2}_{\text{effective}} = \frac{k^2}{\frac{1}{\beta^2\Delta^2}+1} \)
- \( \Delta^2 = \frac{\varepsilon KT}{q^2 n_o} \)

- \( \varepsilon \) – permittivity
- \( \beta \) – acoustic wavenumber
- \( K \) – Boltzmann’s constant
- \( q \) – electron charge
- \( n_o \) – mobile carrier density
Piezoelectric Screening Effect

- Increase in dielectric loss is consistent with decrease in pulse Q

- Decrease in pulse Q is inconsistent with degradation of coupling to propagation medium