Effects of High Energy X Ray and Proton Irradiation on Lead Zirconate Titanate Thin Films

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Introduction

- Ferroelectric thin films with large dielectric permittivity, piezoelectric coefficients and electromechanical coupling are of important for numerous applications:
  - random access memories
  - multilayer capacitors
  - energy harvesting devices
  - micro- and nano-electromechanical sensors and actuators[1,2]
- Especially of interest are autonomous microsystems operating in locations with high radiation exposure such as:
  - nuclear power plants
  - space applications

Research Objective:

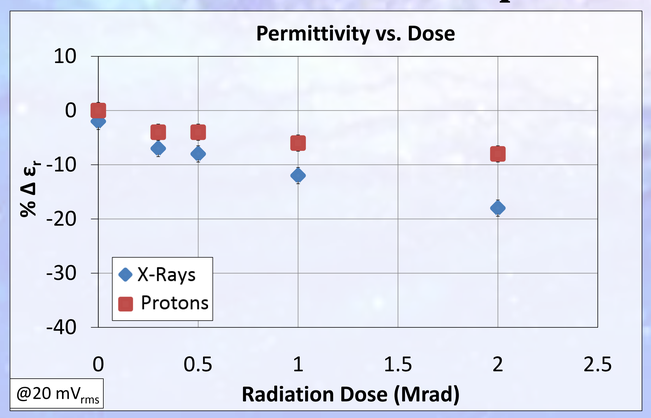
- Investigate effects of irradiation on the dielectric and piezoelectric response of:
  - highly (100)-textured
  - 810 nm thick
  - $Pb(Zr_{0.53}Ti_{0.47})O_3$ (PZT)

  thin films as a function of irradiation by X rays and protons.
- All measurements were performed at 1kHz.

1 Mrad	2 Mrad
0.5 Mrad	0 Mrad

Irradiation Source	Energy
Protons	3 MeV
X rays	10 keV

Low Field Dielectric Response



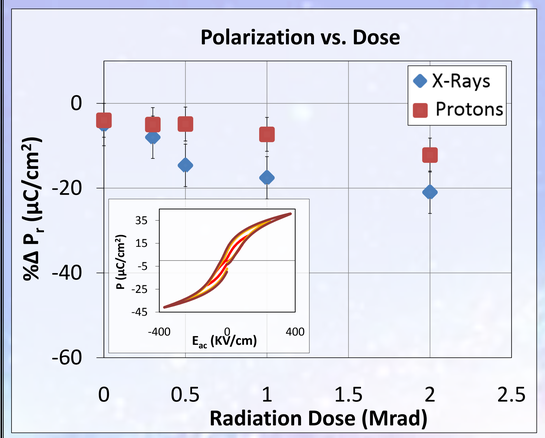
Reference Electrodes	
low-field relative permittivity (ε_r)	1030 ± 30
dissipation factor (tan δ)	1.2% ± 0.2%

- Low field dielectric permittivity decreased with increased radiation.
- Dissipation factors for all samples showed a reduction of less than 0.3% after exposure, independent of the dose.
- A slight reduction in ε_r was also observed in the reference electrodes, which possibly owing to exposure to small radiation due to imperfect coverage.

References

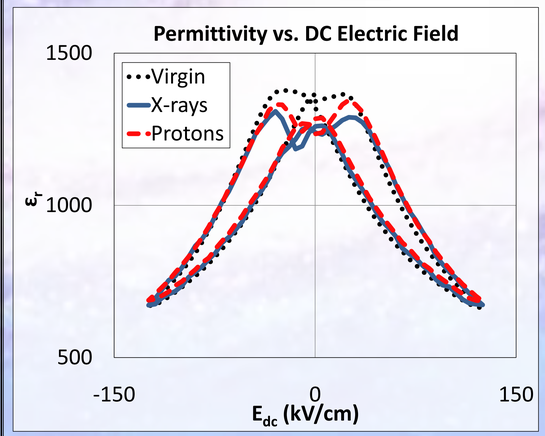
[1] P. Muralt, R. G. Polcawich, and S. Trolier-McKinstry, MRS Bull. 34, 658–66a4 (2009).
[2] G. L. Smith, J. S. Pulskamp, L. M. Sanchez, D. M. Potrepka, R. M. Proie, T. G. Ivanov, R. Q. Rudy, W. D. Nothwang, S. S. Bedair, C. D. Meyer, and R. G. Polcawich, J. Am. Ceram. Soc. **95**(6), 1777–1792 (2012).
[3] J. M. Bendetto, R. A. Moore, F. B. McLean, and P. S. Brody, IEEE Trans. Nucl. Sci. **37**(6), 1713–1717 (1990).

High-Field Dielectric Response:



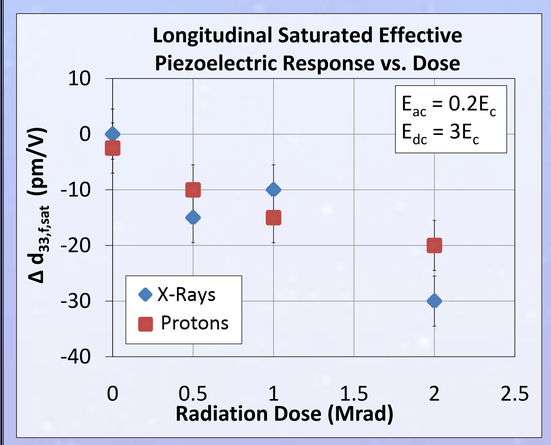
- Reduction in remanent Polarization (P_r) with increasing radiation dose.
- Implies a degradation of ferroelectric switching capability.
- Pinching features were observed in the irradiated samples' minor hysteresis loops.
- These are usually associated with the presence of defect dipoles (pining centers) in the films.[3]

Capacitance-Voltage



- Appearance of additional peaks in C-V curves after irradiation.
- These peaks are associated with the presence of:
  - more than one polarization switching mechanism.
  - substantially different switching activation energies.
- Additional peaks can be associated with new families of pinning sites, i.e. radiation induced-defects.

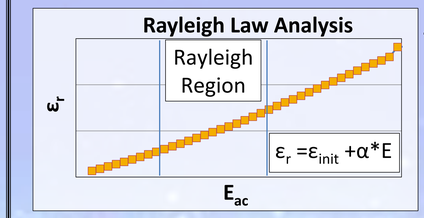
Piezoelectric Response ($d_{33,f}$)



- Note: E_c = coercive field

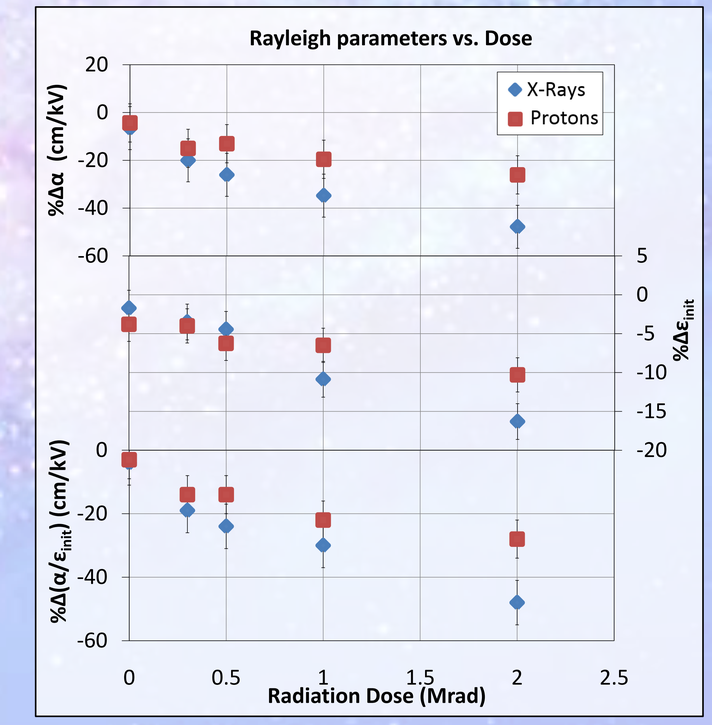
- The converse, effective longitudinal piezoelectric response (d_{33f}) decreases with increasing irradiation dose.
- Indicates that radiation-induced point defects are acting as pinning centers for ferroelastic domain wall motion.
- Reduction of extrinsic electromechanical contributions by irradiation.

Extrinsic Dielectric Response



- Rayleigh law analysis applied to ferroelectric materials allows quantification of extrinsic contributions of domain walls and phase boundaries (interacting with pinning centers).

ε_{init}: lattice and reversible extrinsic contributions
$\alpha * E$: irreversible extrinsic contributions
$\alpha / \varepsilon_{init}$: ratio of irreversible to reversible Rayleigh parameters



- At increasing radiation dose (up to 2Mrad) the films showed a decrease of:
  - α by 50%
  - ε_{init} by 15%
  - $\alpha / \varepsilon_{init}$ by 40%

  → Large reduction of extrinsic dielectric contributions.
- Consistent with increase of radiation-induced pinning centers (point defects) for domain wall motion.
- A post-radiation heat treatment above the Curie temperature results in almost full recovery (within ±3%) of the dielectric response.
- Consistent with annealing of point defects.

Conclusions

- Low field dielectric permittivity, remanent polarization, and piezoelectric $d_{33,f}$ response degraded with exposure to radiation.
- Dielectric and piezoelectric characterization suggest a radiation-induced reduction of the extrinsic contributions to the response, attributed to increased pinning of the domain walls by the radiation-induced point defects.
- The response is recoverable within 3% of the original properties via a post-radiation heat treatment at above Curie temperature.