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Exploring the evolution of the polarization behavior for Nb-PZT based on the electric field-pressure conditions

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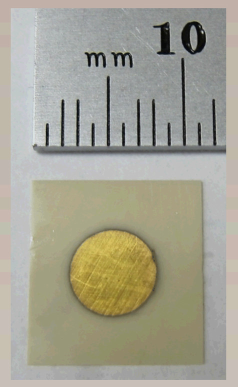
MOTIVATION

Lead zirconate stannate titanate ceramic ($Pb(Zr,Sn,Ti)O_3$) is an important family of ferroelectrics and is usually modified by niobium (Nb) or lanthanum (La) to tailor its properties. In particular, compositions close to the phase boundary of the ferroelectric (FE) and antiferroelectric (AFE) have attracted considerable interest for many years. Near the FE/AFE phase boundary, the free energy difference between the FE and AFE phase is very small, indicating that the FE/AFE phase transition can occur easily with an applied electric field or pressure. These phase transitions are usually accompanied by a volume expansion/contraction, a development/release of electrical polarization, or an incommensurate modulation in structure, resulting in the possibility for many engineering applications.

Niobium modified lead zirconate titanate ceramic with a Zr:Ti ratio of 95:5 (95/5 PZT) undergoes a hydrostatic pressure induced phase transformation from rhombohedral ferroelectric to orthorhombic antiferroelectric. In the FE state, electric fields can induce a remnant polarization due to domain restructuring within the material. During the phase transformation from the FE to the AFE state, the remnant polarization goes to zero. This study will present work on experimental measurements of the effect of combined bipolar electric field cycles and hydrostatic pressure on the FE-AFE phase transformations in various formulations of niobium modified $Pb(Zr,Ti)O_3$. Specifically investigating the evolution of the hysteresis looping behavior as a function of the applied hydrostatic pressure. The motivations for this work include; development of an improved understanding of the electric-field dielectric displacement behavior of the FE-AFE phase transformation, potential for construction of phase diagrams for electric field-pressure conditions, and the possibility for an improved understanding of the physics of the FE-AFE phase transformation for use in the creation of phase transforming material models.

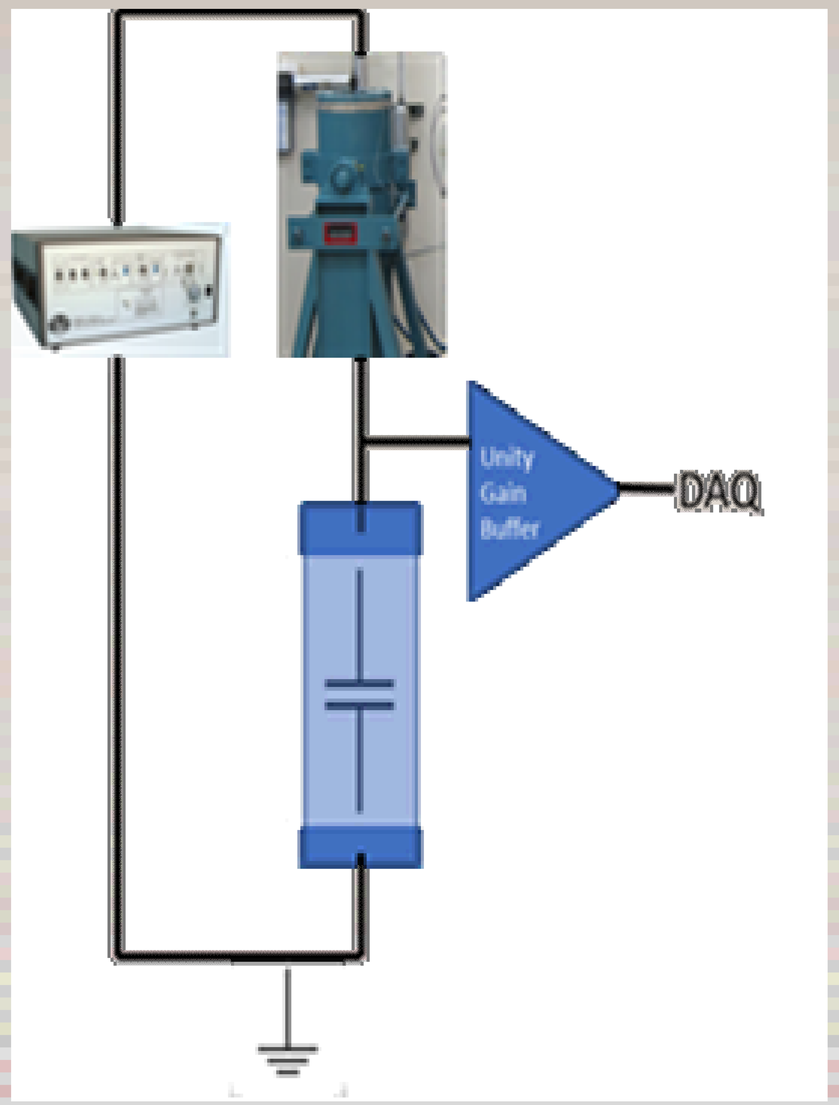
TEST SAMPLE CONFIGURATION

- The samples used for this study were approximately 1cm x 1cm x 0.022cm (220μm) in physical dimensions and were prepared with a minimum thickness to balance the mechanical integrity of the sample and to maximize the applied electric field based on the high voltage limitations of the feedthroughs on the seal to the high-pressure vessel.
- A recessed gold sputtered electrode, approximately 6mm in diameter, was applied to the center of each of the large faces of the samples resulting in an area of approximately $0.282 cm^2$.



HYSTERESIS LOOP – HYDROSTATIC PRESSURE TEST

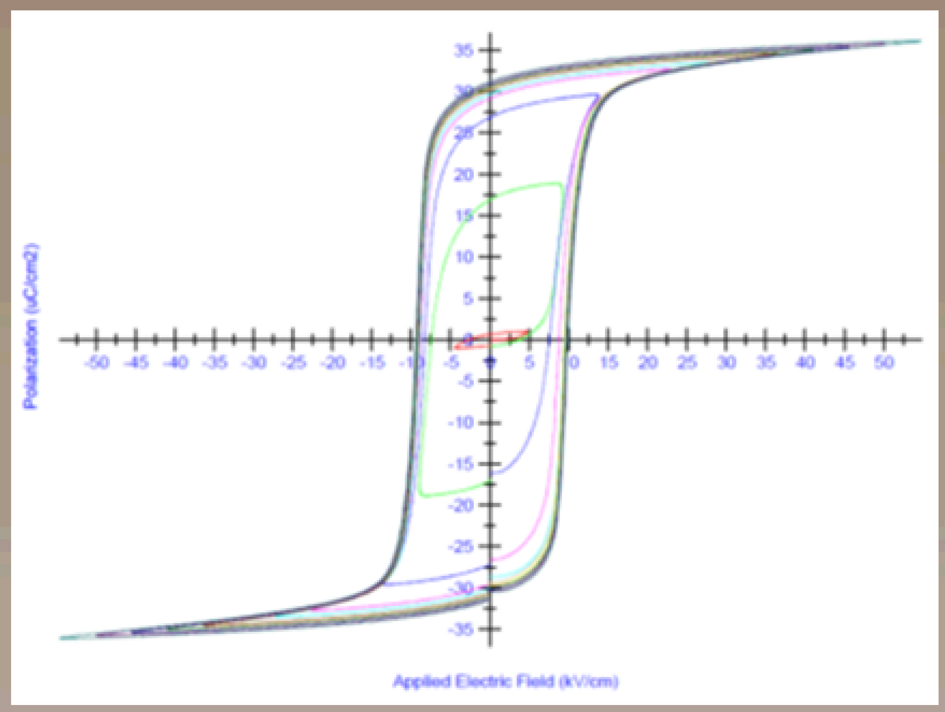
- A custom designed and built hysteresis looping-hydrostatic depoling test system was used to collect and characterize the hysteresis loop behavior as a function of hydrostatic pressure.
- The pressure vessel (Harwood Engineering Company, Walpole, MA) has a 2" diameter, 8" long cavity containing a custom Teflon sample holder and copper battery tabs for electrical connections.
- Pumping system, single-acting intensifier, that will pressurize the vessel in one stroke at controllable rates (Monoplex DOS (diethyl ethyl sebacate).
- The system pressure, the applied electric voltage (TREK amplifier) and any electrical measurements (high voltage buffer/unity gain amplifier (unity gain buffer), integrating capacitor) are controlled and monitored through a programmable controller that is operated from a LabVIEW based data acquisition system.
- The hysteresis loop behavior was measured as a function of applied hydrostatic pressure during a continuous, slow-rising pressure ramp (approximately 250 PSI/second).
- Based on the overall measurement sampling frequency, hysteresis loops were collected every 750 PSI, with approximately 100 PSI of ambiguity).



HYSTERESIS BEHAVIOR

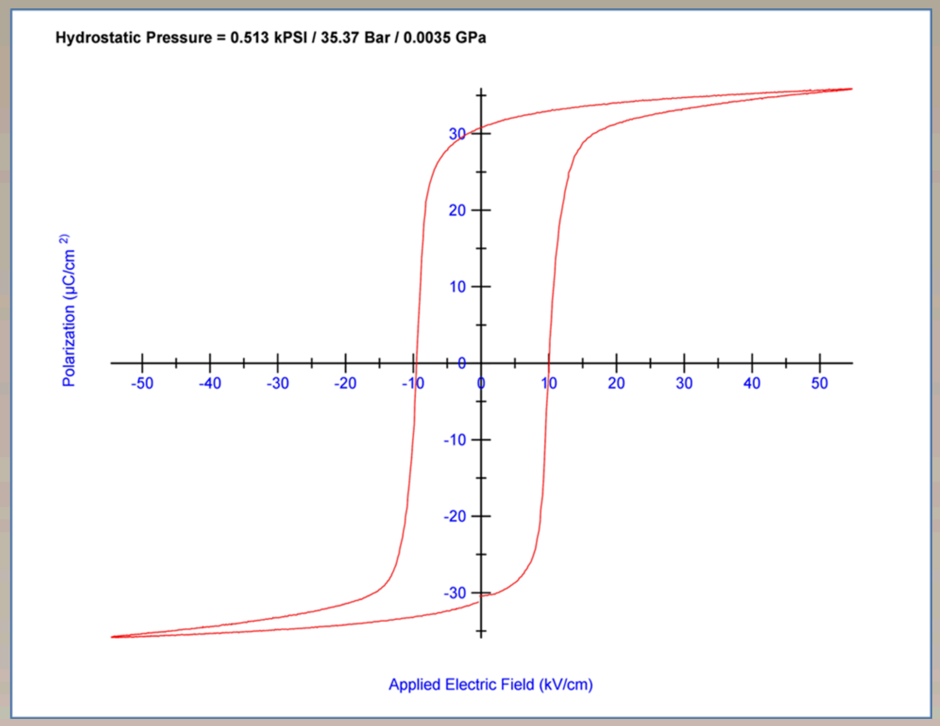
HYSTERESIS BEHAVIOR – AMBIENT CONDITIONS

- The hysteresis loop behavior of the sample under test in a static condition (in the hydrostatic pressure vessel, ambient temperature and pressure) were initially measured as a function of applied electric field
- These measurements permitted the determination of a maximum allowable voltage estimation and verified that the test circuit was functional and did not introduce any additional loss of significance.
- The hysteresis loop behavior was then captured from ambient pressure to approximately 60kPSI using a 55 kV/cm applied electric field every 750 PSI.



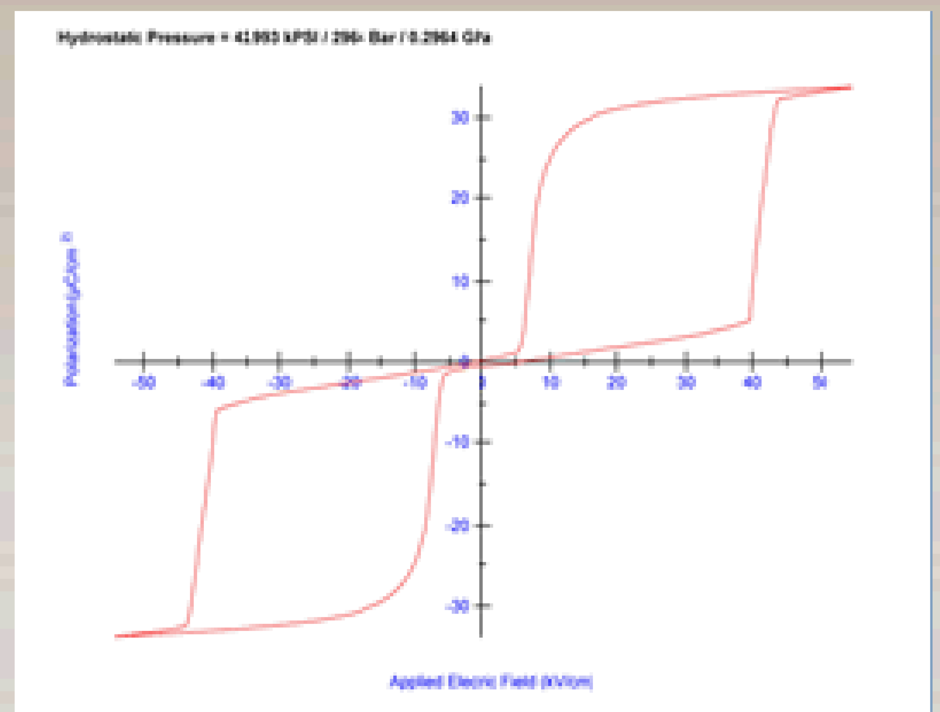
HYSTERESIS BEHAVIOR – FE PHASE

- The hysteresis loop immediately following the initiation of the hydrostatic pressure cycle at 0.5 kPSI, 35.37 Bar, 3.5 MPa.
- At this low pressure, below the FE-AFE phase transformation pressure, the shape of the polarization-applied electric field takes on the characteristic single loop shape, typical of the FE phase.
- The shape is based on the FE phase retaining remnant polarization that is only reoriented when the electric field reaches the coercive field.



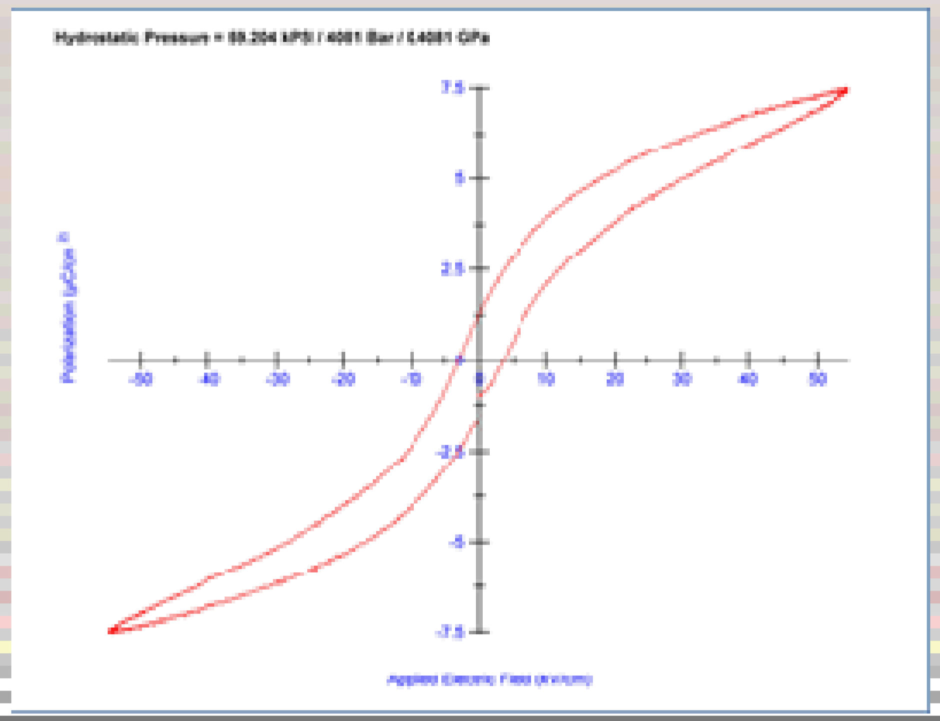
HYSTERESIS BEHAVIOR – AFE-FE PHASE

- Hysteresis loop during the hydrostatic pressure cycle at 43 kPSI, 2964 Bar, 296 MPa.
- At this pressure condition the material is in the "double hysteresis loop regime".
- A non-hysteretic AFE region occurs between the two separated hysteresis loops.



HYSTERESIS BEHAVIOR – AFE PHASE

- As the pressure is further increased, the hysteresis loop during the hydrostatic pressure cycle at 59 kPSI, 4081 Bar, 408 MPa, the applied electric field is now insufficient to drive the AFE to FE transformation and the hysteresis loop takes on a non-hysteretic dielectric behavior.



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