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Textured Processing, Reactive Templated Grain Growth, and Electrical Property Relationships for Sodium Bismuth Titanate

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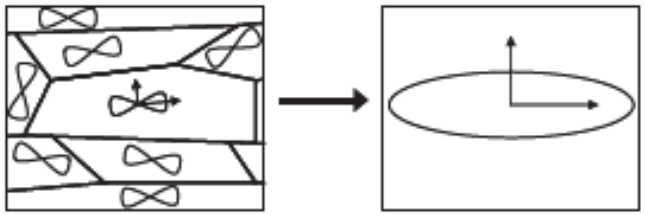
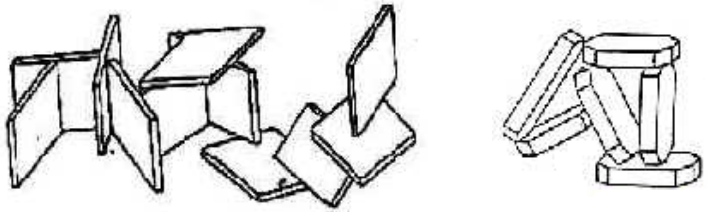
Microstructural engineering through processing induced texturing

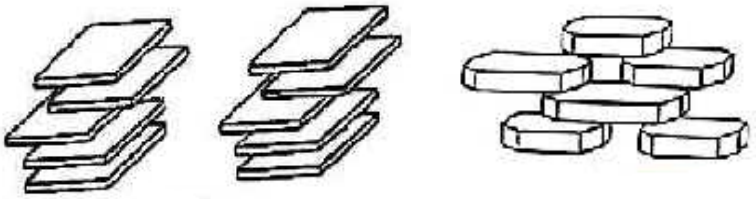
- Target – Develop an advanced, low cost manufacturing process for a non-lead based microstructural engineered ferroelectric ceramic material
  - Microstructural engineering through texture modifications
    - Template induced texturing process for a bulk ceramic
- Coupled crystal orientation and domain orientation processing yields ceramics with enhanced macroscopic properties
- Influence of the micro-anisotropy of the crystals on the macro-anistropy of the material



- **House of Cards**
  - Random Grain Orientation



- **Deck of Cards**
  - Preferential Grain Orientation



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J.L. Jones, "Texture and Anisotropy of Polycrystalline Piezoelectrics", JACERS, 90 [8] 2297-2314 (2007)

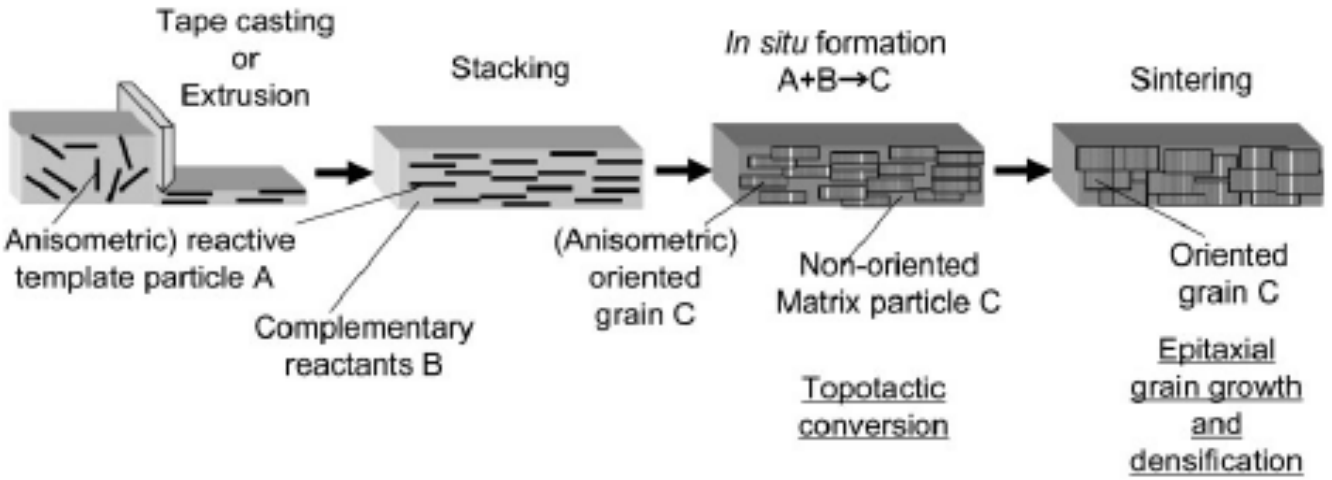
Templated Grain Growth vs Reactive Templated Grain Growth

- Templated Grain Growth (TGG)
  - Templates embedded in the ceramic matrix – nominally the same composition
    - Anisometric particles mixed with other fine equiaxed particles
  - Heat treatment conditions are key factors in the growth process of the textured ceramic
  - Difficult to prepare anisometric template particles with a pseudo-cubic simple perovskite type structure of the target composition

- Reactive Templated Grain Growth (RTGG)
  - Reaction sintering based technique
  - Anisometric particles - simpler composition and easier fabrication route than the target material
  - Particles are used as precursors to be aligned and converted into the target material
  - Preserve the crystallographic orientation of the templates

Project Overview and Background

- Fabrication of a "Functional" Material via a Novel Process
  - Powder synthesis
    - Mixed oxide technique
  - Synthesis of templated seed crystals
    - Morphologically controlled templates
    - Preferably oriented polycrystals
    - Fused salt or molten salt synthesis technique
  - Texture Engineering - Mutual orientation of the crystallographic lattices of the grains.
    - TGG or RTGG methods
    - Texture development – highly localized shear
      - Induced shear stresses result in particle alignment - preferred stress direction



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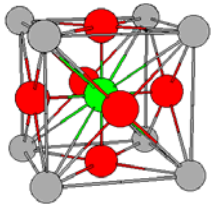
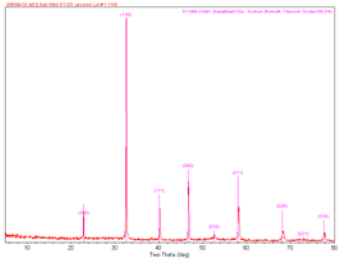
T. Tani and T. Kimura, "Reactive-templated grain growth processing for lead free piezoelectric ceramics", Adv. in Appl. Ceram., 105 [1] (2006)

$Bi_4Ti_3O_{12}$ (BTO)–$BaTiO_3$ (BT)–$Na_{1/2}Bi_{1/2}$ TiO_3 (NBT)

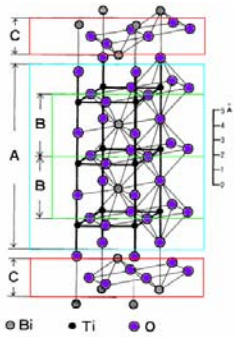
Phase pure $(Na_{0.5}Bi_{0.5})TiO_3$

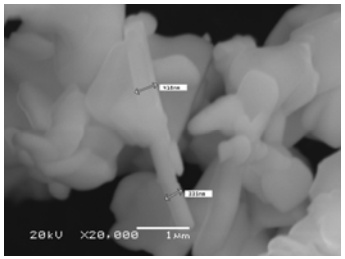
Space group: Pm-3m
Lattice parameters:
a = 3.878(2) Å
Cell volume = 58.32 Å^3

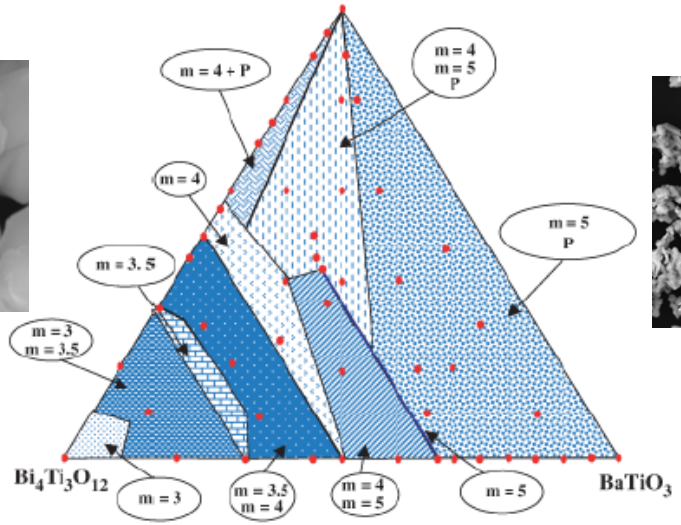


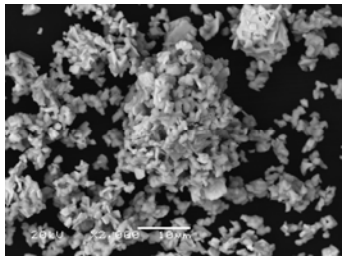


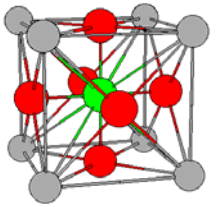
$Na_{0.5}Bi_{0.5}TiO_3$





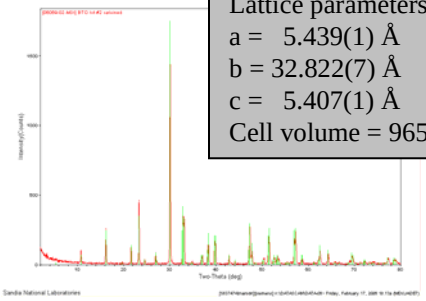


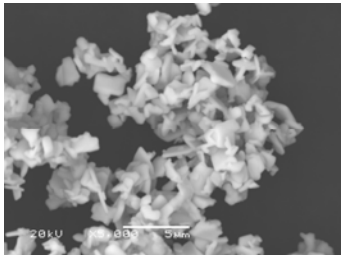


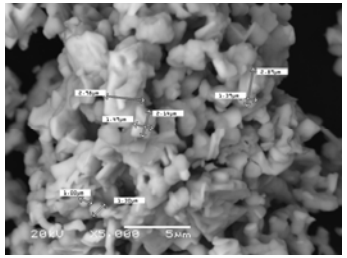


Phase pure $Bi_4Ti_3O_{12}$

Space group:Cmmm
Lattice parameters:
a = 5.439(1) Å
b = 32.822(7) Å
c = 5.407(1) Å
Cell volume = 965.3 Å^3

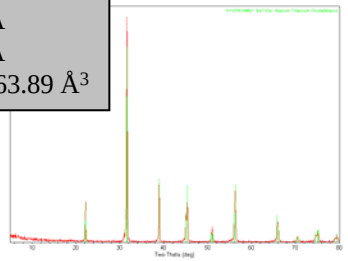






Phase pure $BaTiO_3$

Space group:P4/mmm
Lattice parameters:
a = 3.989(1) Å
c = 4.015(1) Å
Cell volume = 63.89 Å^3



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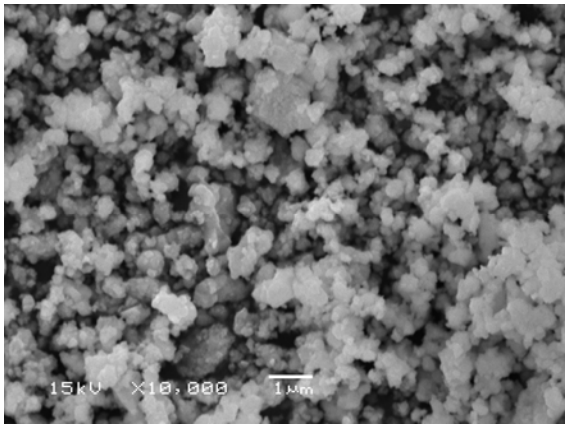
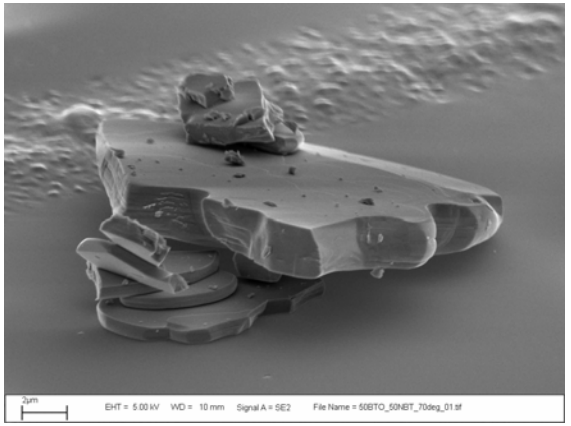
Material and Sample Synthesis

- Powder Synthesis – $Na_{0.50}Bi_{0.50}TiO_3$:
  - Mixed Oxide Solid State Reaction – Na_2CO_3, Bi_2O_3, and TiO_2
  - Ball milled – Y_2O_3-ZrO_2 milling media
  - Freeze Dried
  - Calcined 850^{o}C for 2hours
  - Jet milled or attritor milled



- Template Seed Synthesis - $Na_{0.50}Bi_{4.50}Ti_4O_{15}$
  - Molten Salt Synthesis Method – 50/50 wt% KCl-NaCl
  - Thermal treatment - sealed in platinum crucibles – washed and filtered

- Sample Preparation
  - Pellets
    - PVA/PEG Binder system
    - Dry Pressed – Isostatic Pressed to 30kPSI
    - Sintered to 1225oC for 2-4 hours
    - Test samples polished and sputtered with Pt electrodes
  - Tape cast laminates
    - Ferro B73305 tape cast vehicle, Hypermer KD1, toluene
    - 70wt% solids total, templated seeds – 5-30wt%
    - Ball milled, 3-roll milled, de-aired and cast
    - Wet tape thickness = 127um, dry tape thickness = 50-55um
    - Laminated layers at 65-75oC
    - Sintered to 1225oC for 2-4 hours, polished and Pt-sputtered electrodes



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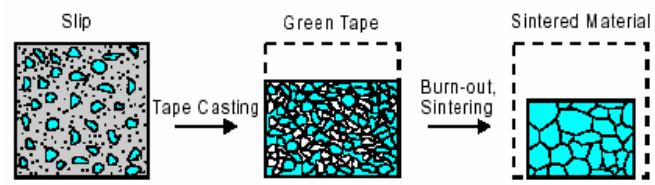
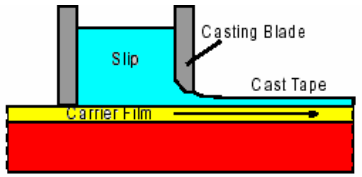
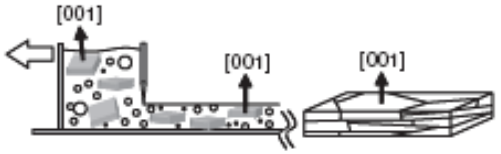
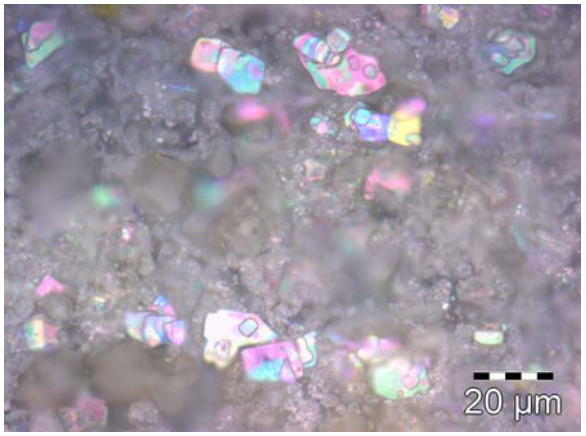
Textured microstructures are developed through shear-inducing forming techniques

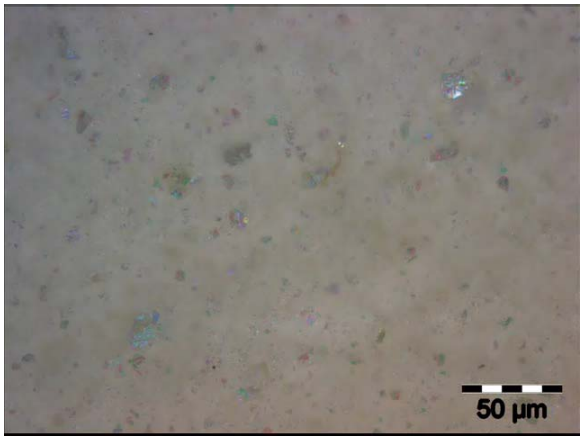
- Tape casting and screen printing are used to develop a textured microstructure
- Powder matrix is mixed with a seed crystal
- Forming parameters continue to be optimized

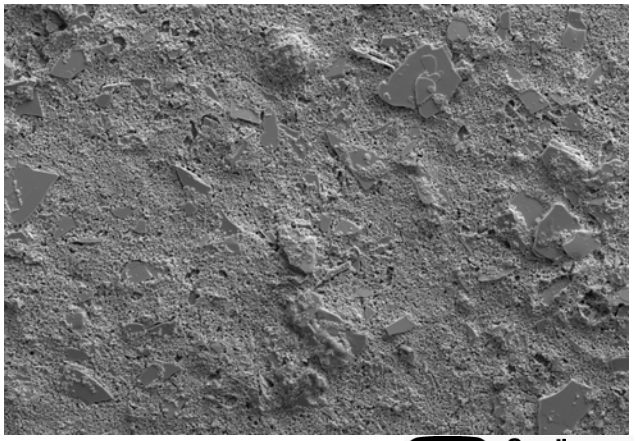












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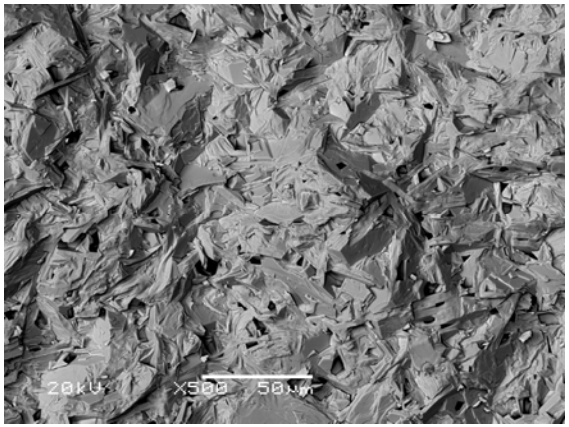
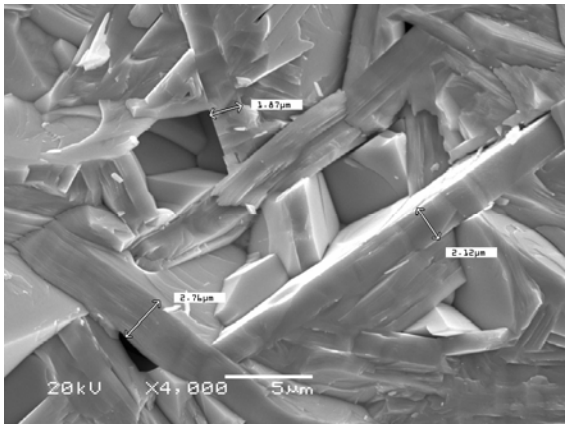
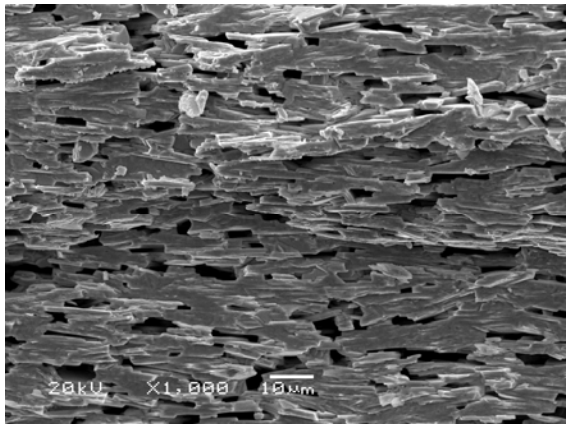
Progression from random to textured dense polycrystalline ceramic microstructures

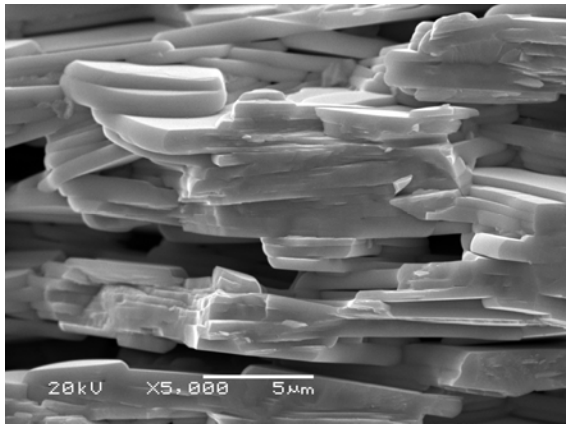
- Random polycrystalline dense microstructure



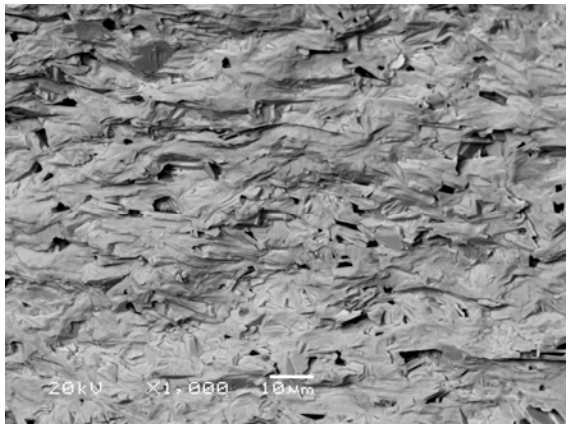


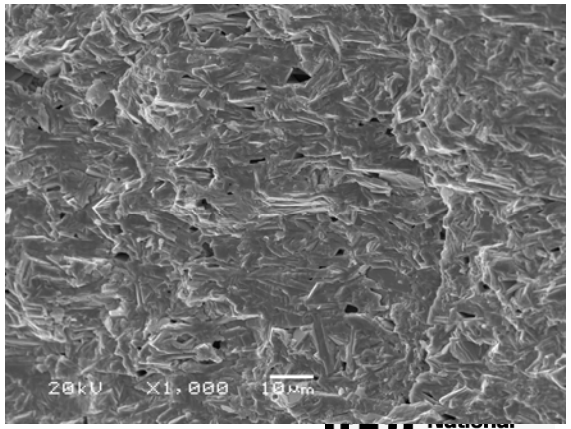
- Textured polycrystalline low-density microstructure





- Textured polycrystalline dense microstructure





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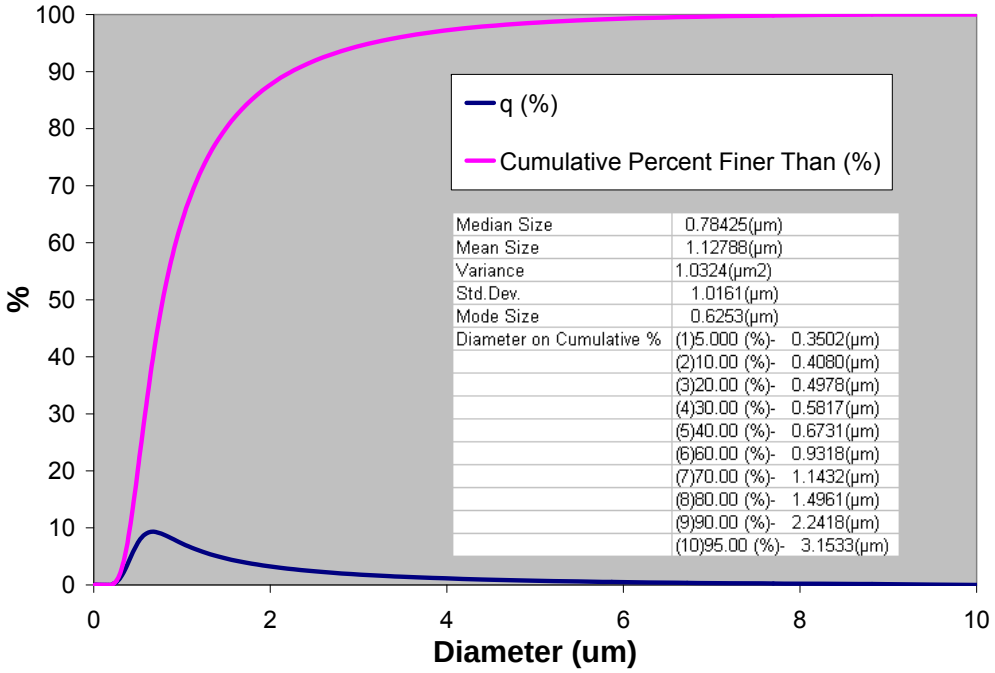
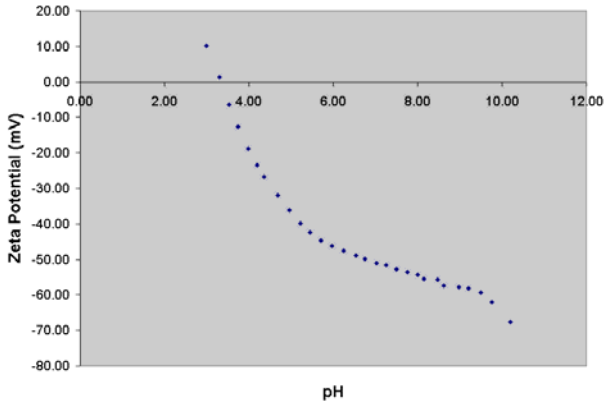
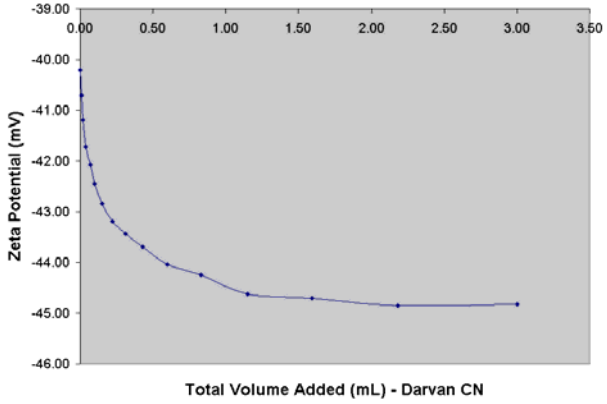
Powder and Colloidal Properties

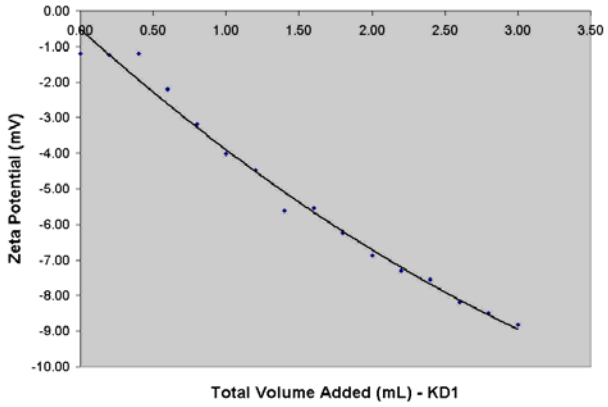
- Powder Characteristics:
  - Density = 5.8926±0.0005g/cm^3
  - BET Multipoint Surface Area = 4.231±0.0149m^2/gram
  - Laser Scattering PS ≈ 1.13um
  - Zeta Potential Analysis – Water / Toluene



Median Size	0.78425(µm)
Mean Size	1.12788(µm)
Variance	1.0324(µm2)
Std.Dev.	1.0161(µm)
Mode Size	0.6253(µm)
Diameter on Cumulative %	(1)5.000 (%)- 0.3502(µm)
	(2)10.00 (%)- 0.4080(µm)
	(3)20.00 (%)- 0.4978(µm)
	(4)30.00 (%)- 0.5817(µm)
	(5)40.00 (%)- 0.6731(µm)
	(6)60.00 (%)- 0.9318(µm)
	(7)70.00 (%)- 1.1432(µm)
	(8)80.00 (%)- 1.4961(µm)
	(9)90.00 (%)- 2.2418(µm)
	(10)95.00 (%)- 3.1533(µm)







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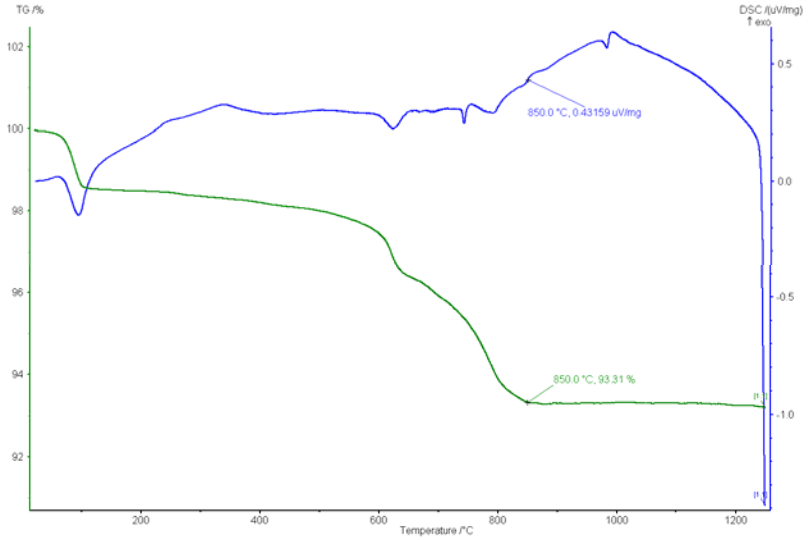
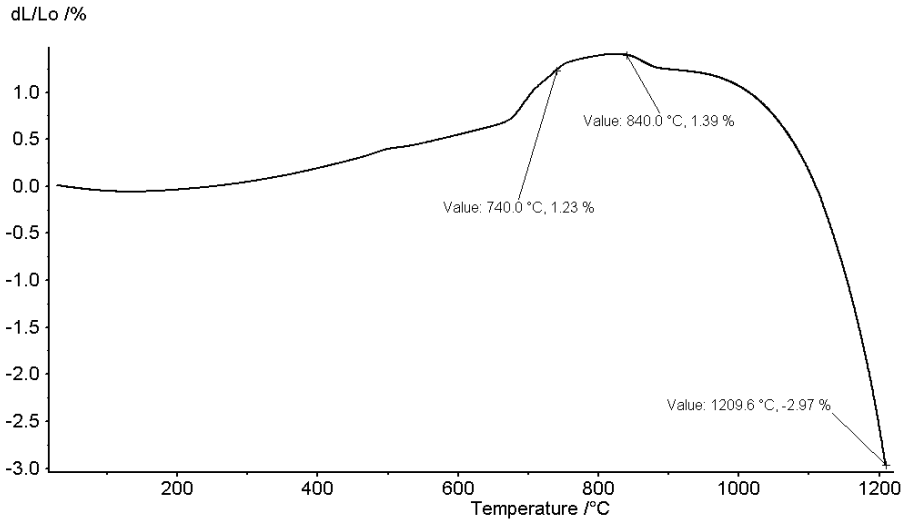
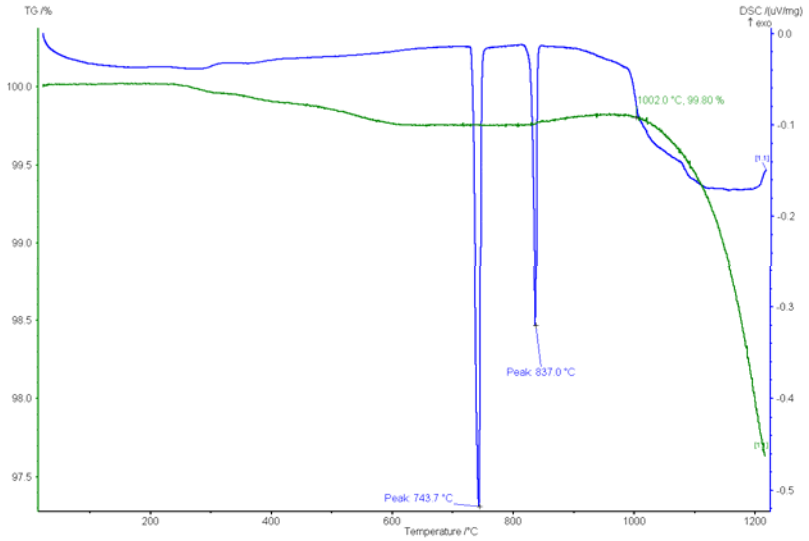
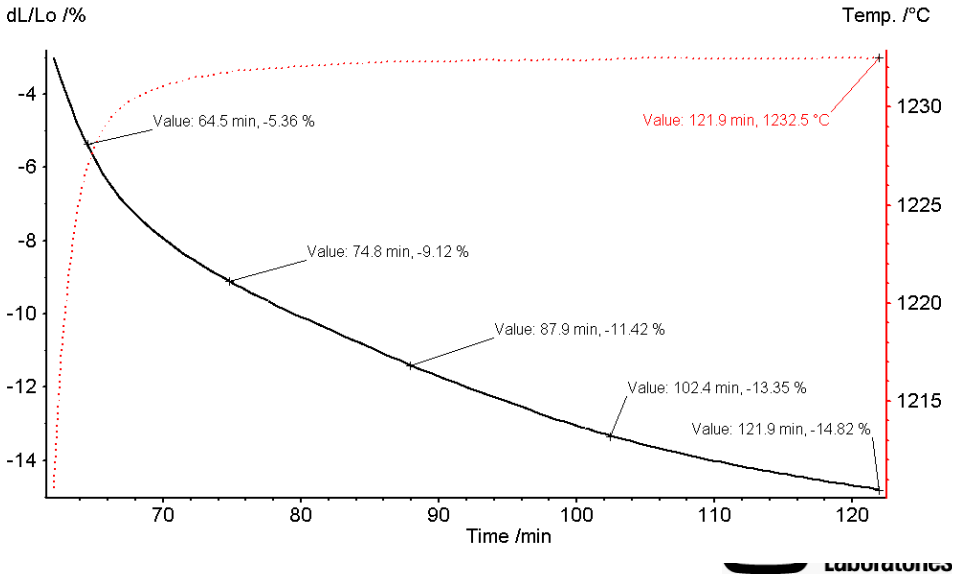
Thermophysical Characterization of NBT



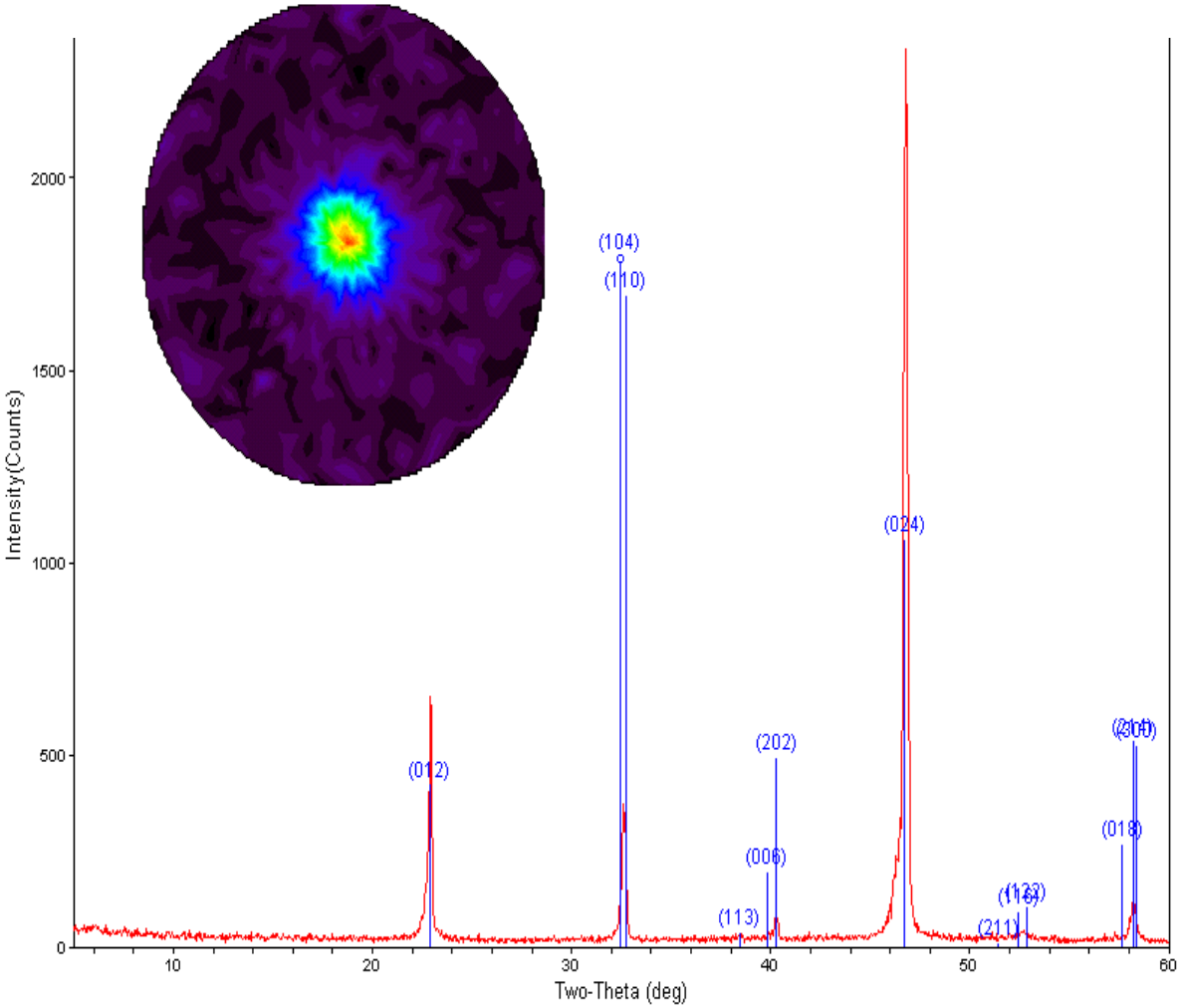






X-ray Diffraction θ-2θ scan and pole figure analysis for templated NBT

- Standard θ-2θ Scan:
  - Material appears to have a (012) preferred orientation
  - Hexagonal setting of the R3c structure
  - Unusual out-of-plane texture was confirmed using pole figure analysis
  - Strong central intensity implies out-of-plane texture for the (012) peak, ≈22.9° 2θ



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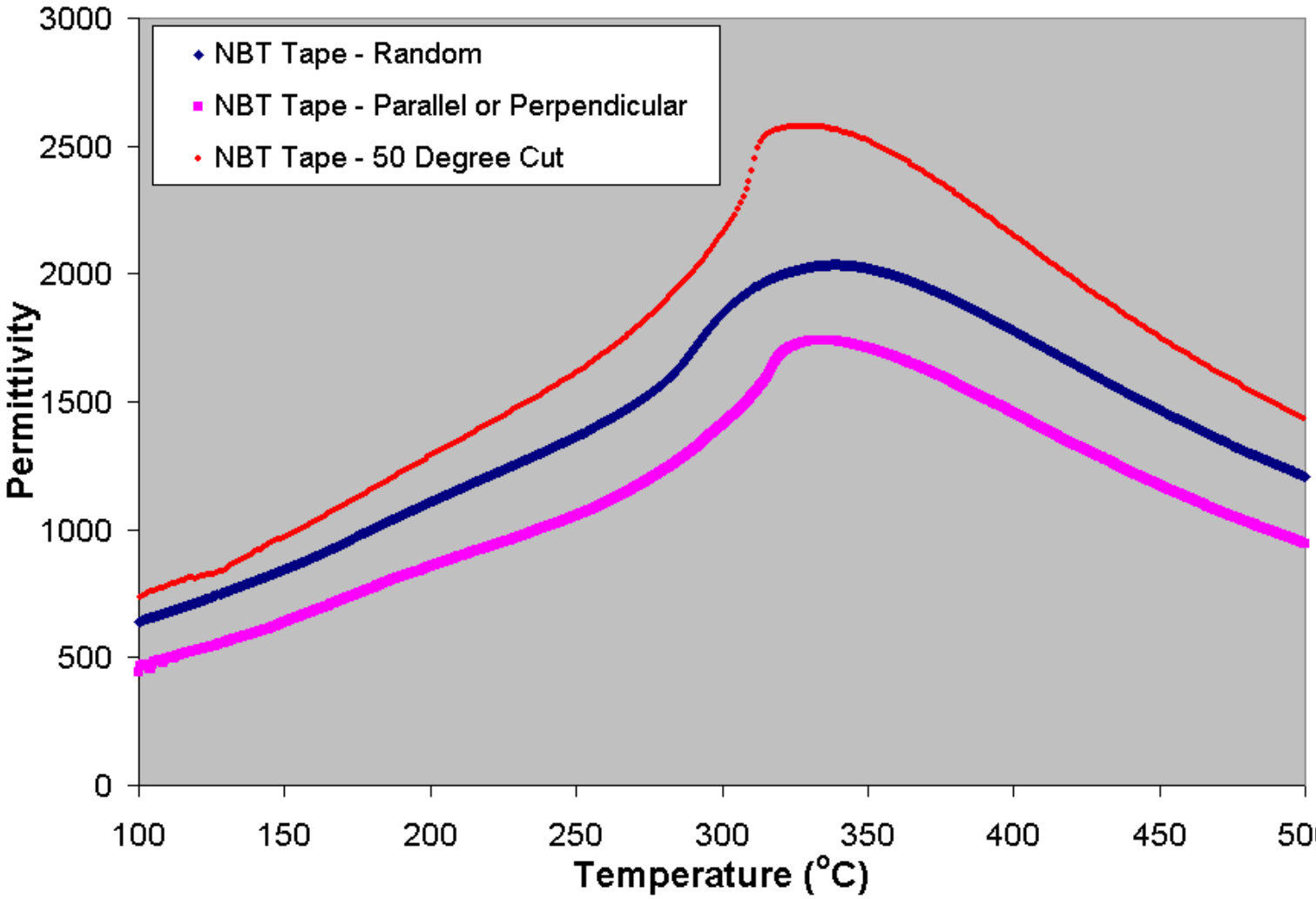
Anisotropic weak-field dielectric behavior

- Weak-Field Dielectric Properties – NBT System



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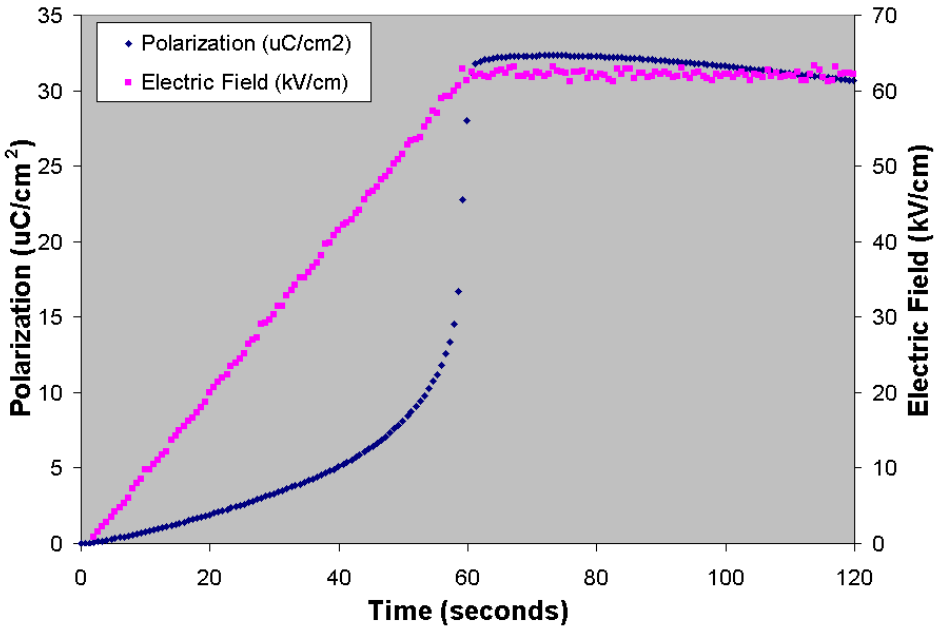
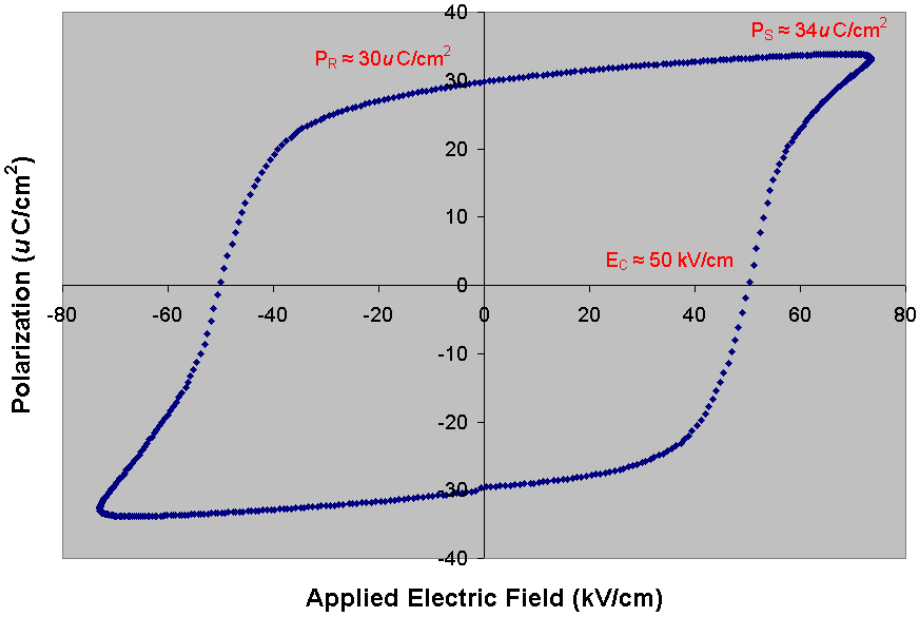
High Field Poling and Polarization Behavior of NBT

- Room Temperature Poling and Hysteresis Loop Behavior
  - 63kV/cm, 2 minutes
  - $P_r \approx 30 uC/cm^2$, $P_s \approx 34 uC/cm^2$, $E_C \approx 50 kV/cm$
  - Substantial loss and at high field “saturation”





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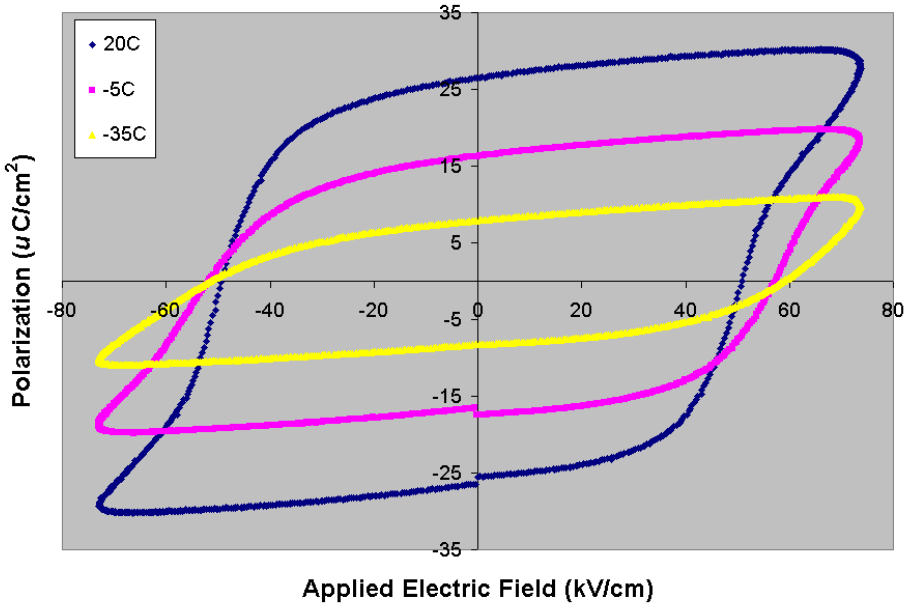
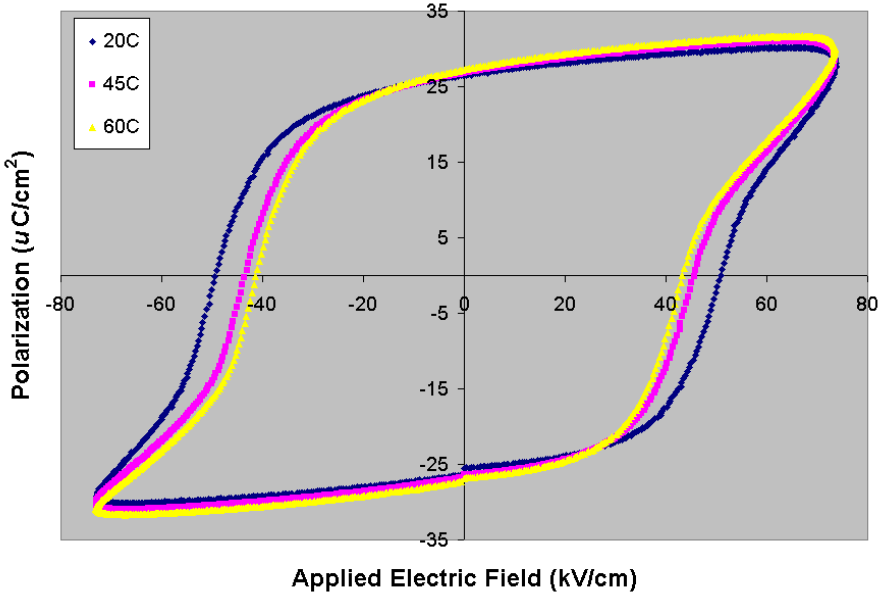
High field polarization behavior as a function of temperature for NBT





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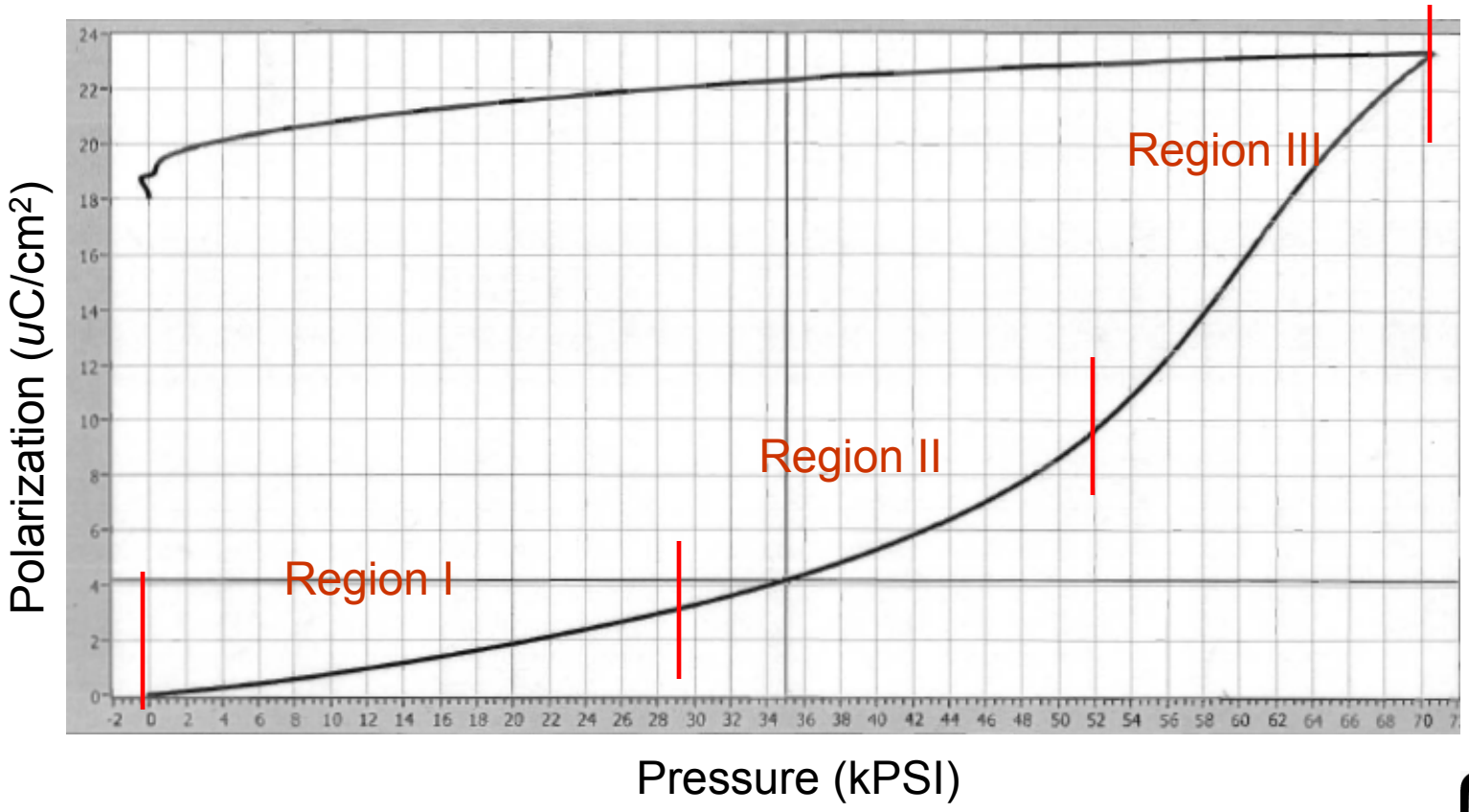
Hydrostatic depoling characteristics for NBT

- Region I = 0.10*u*C/cm^2/kPSI
- Region II = 0.28*u*C/cm^2/kPSI
- Region III = 0.75*u*C/cm^2/kPSI



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