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Enhanced Ferroelectric and Dielectric Property Relationships Induced by Textured Processing for Several Ceramic Compositions

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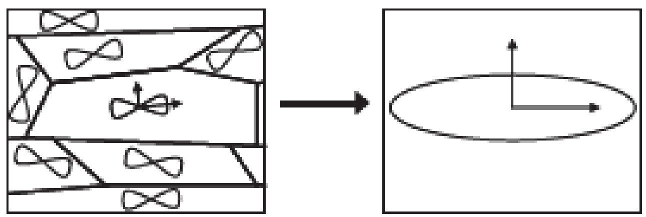
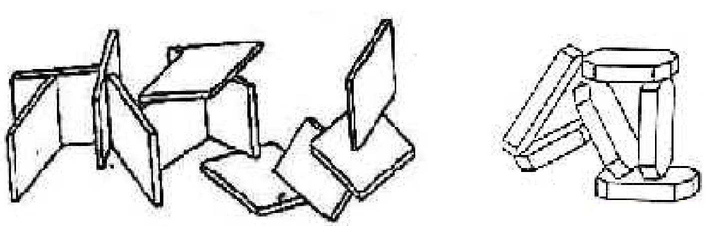
Microstructural engineering through processing induced texturing

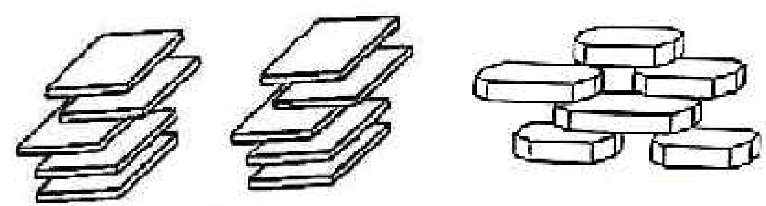
- Target – Develop an advanced, low cost manufacturing process for a non-lead based microstructural engineered ferroelectric ceramic material
  - Microstructural engineering through texture modifications
    - Template induced texturing process for a bulk ceramic
- Coupled crystal orientation and domain 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



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

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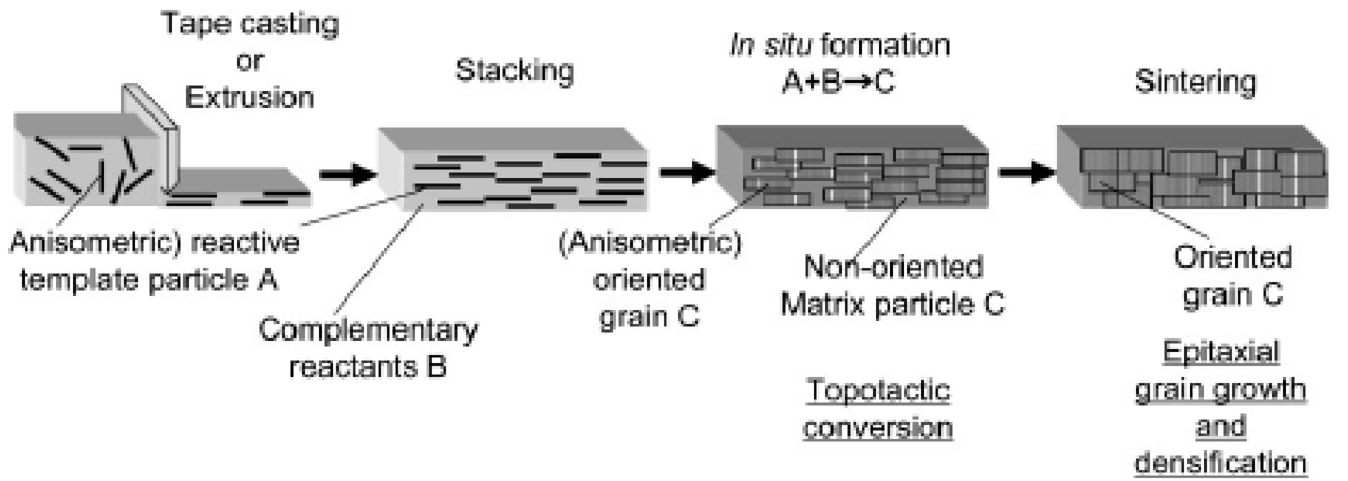
Project Overview and Background

- 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

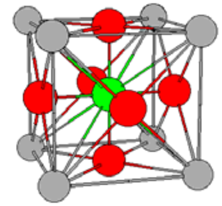


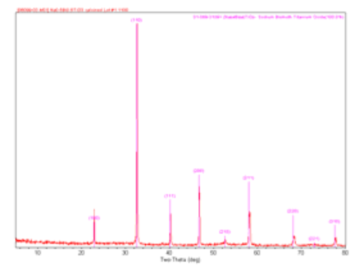
T. Tani and T. Kimura, "Reactive-templated grain growth processing for lead free piezoelectric ceramics", Adv. in Appl. Ceram., 105 [1] (2006)

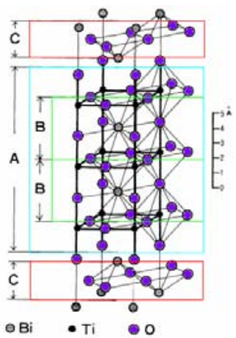
Powder Synthesis - $Bi_4Ti_3O_{12}$ (BTO)-$BaTiO_3$ (BT)-$Na_{1/2}Bi_{1/2}$ TiO_3 (NBT)

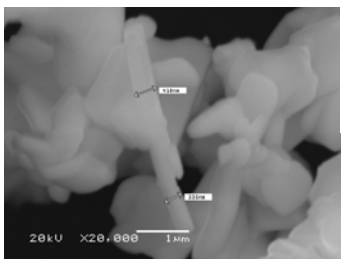
Phase pure $(Na_{0.5}Bi_{0.5})TiO_3$

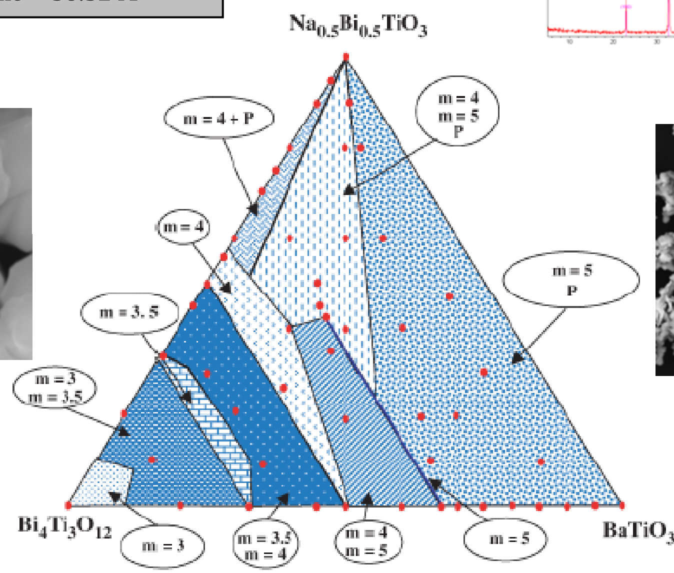
Space group: Pm-3m
Lattice parameters:
a = 3.878(2) Å
Cell volume = 58.32 Å^3

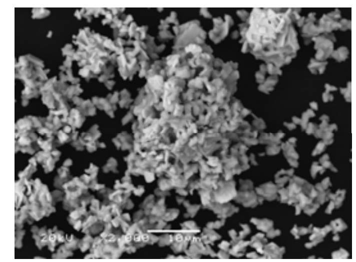


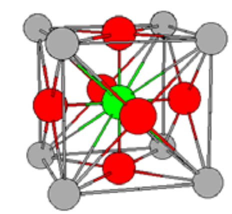










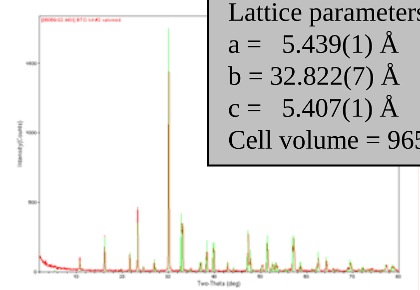


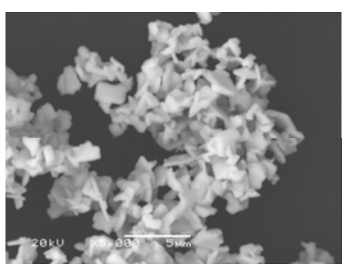
Phase pure $Bi_4Ti_3O_{12}$

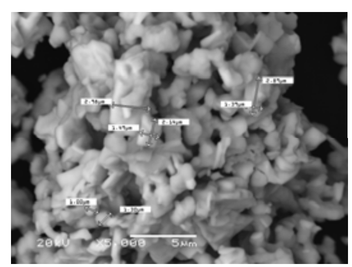
Space group:Cmmm
Lattice parameters:
a = 5.439(1) Å
b = 32.822(7) Å
c = 5.407(1) Å
Cell volume = 965.3 Å^3

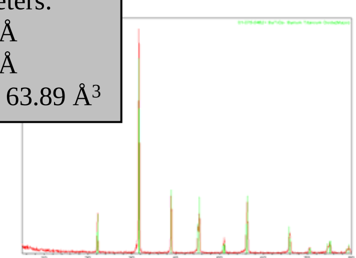
Phase pure $BaTiO_3$

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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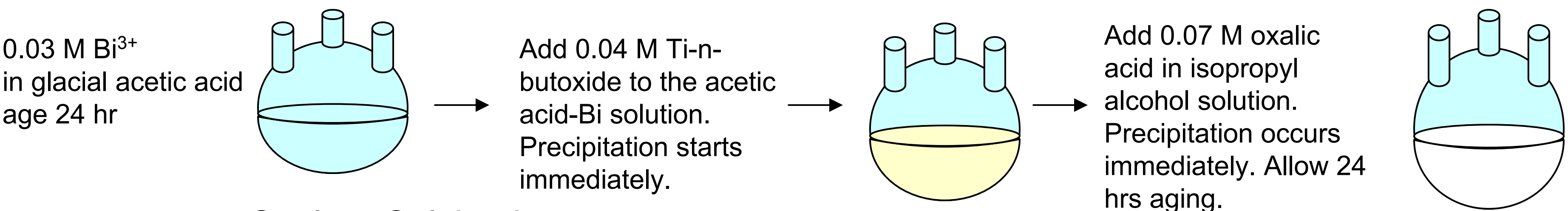
Synthesis of templated seed crystals – chem-prep, mixed oxide and molten salts

- <u>Seed crystal growth technique:</u>

1. Precursor Route:

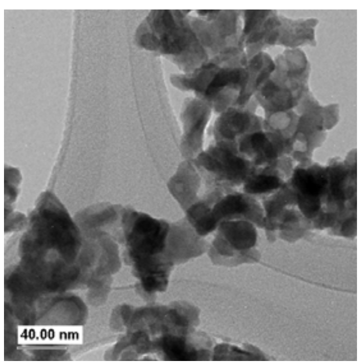
– Synthesize precursor materials – chem-prep procedure

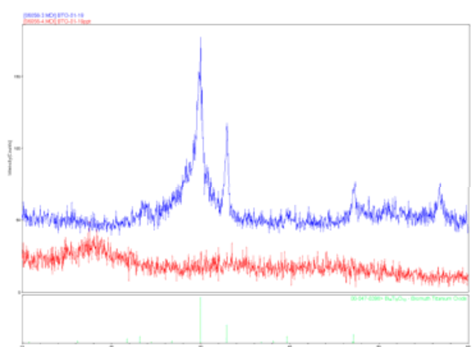


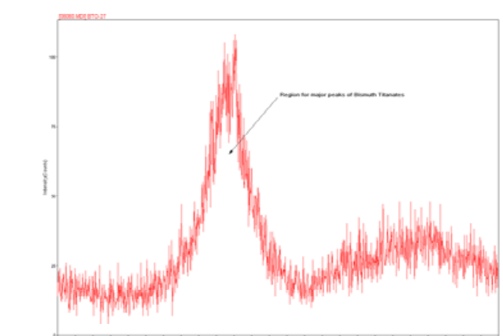
– Oxalate Calcination

  – Bismuth Titanate amorphous precursor

– Precursor molten salt synthesis technique





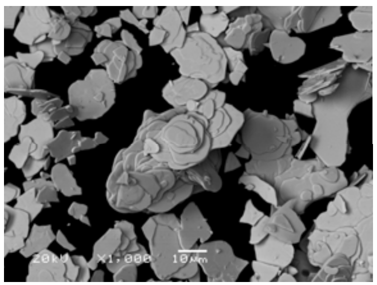


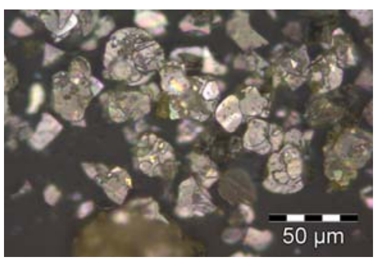
2. Direct Molten Salt Synthesis

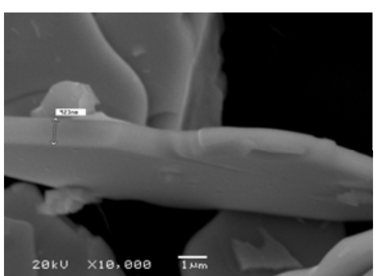
– Oxide + Salt Mixture

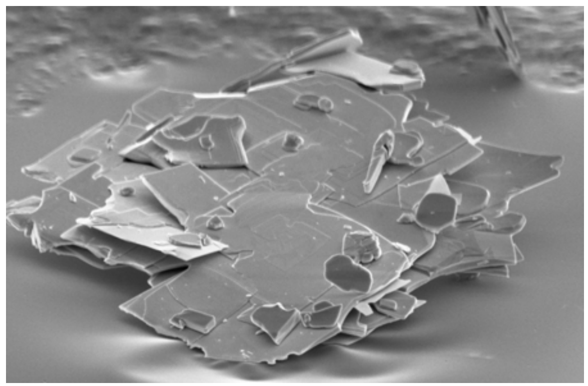
Synthesis of templated seed crystals – chem-prep, mixed oxide and molten salts

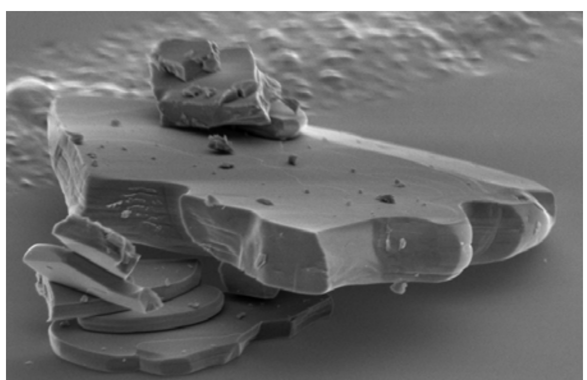
- <u>Seed crystal growth technique:</u>
- $Bi_4Ti_3O_{12}$ - Bismuth titanate (BTO) - express a platelet like morphology
- $Ba_2Bi_4Ti_5O_{18}$ – Barium bismuth titanate - express a platelet like morphology
- $(Na)Bi_4Ti_4O_{15}$ – Sodium bismuth titanate - express a platelet like morphology
- $Sr_{1-x}Ba_xNb_2O_6$ – Strontium barium niobate – express a fiber or whisker like morphology

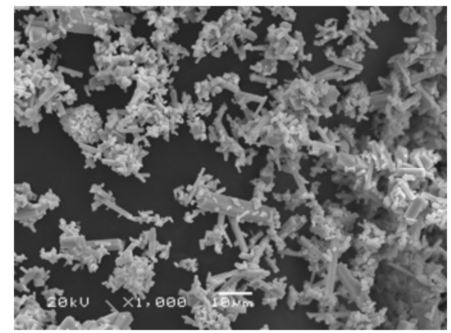


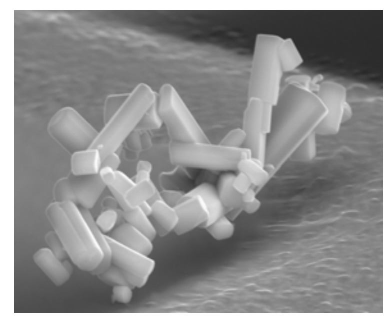


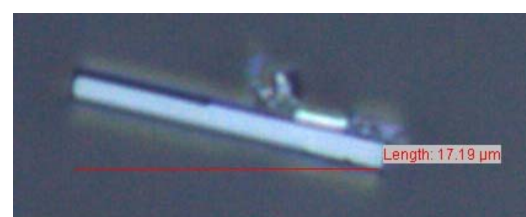












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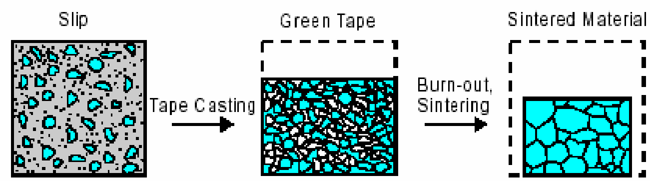
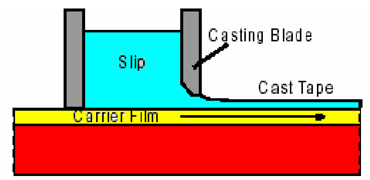
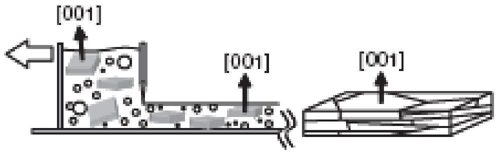
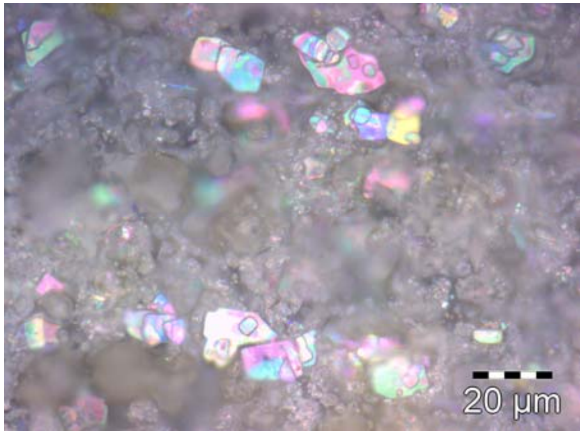
Textured microstructures are developed through shear-inducing forming techniques

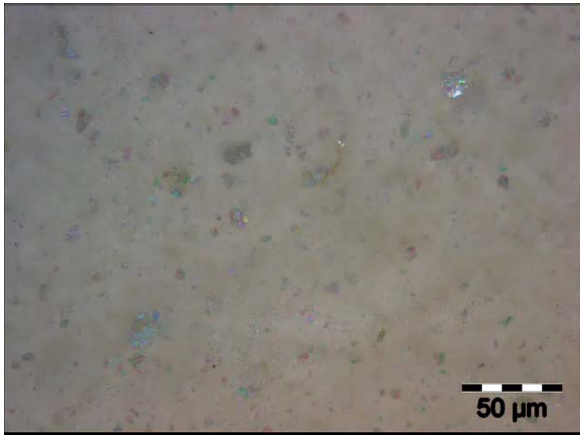
- Tape casting and screen printing are used to develop a textured microstructure
- Powder matrix is mixed with a seed crystal
- Forming parameters are being investigated

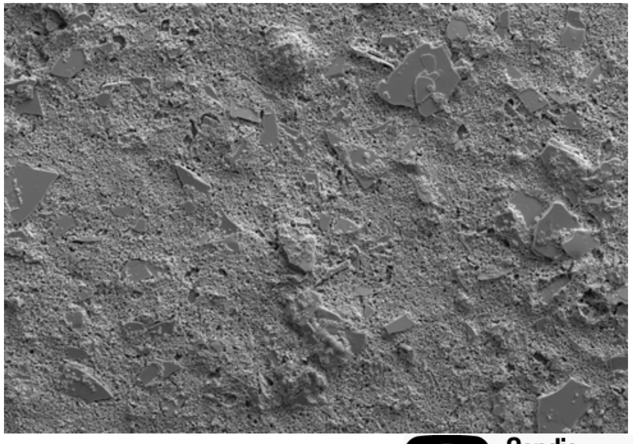












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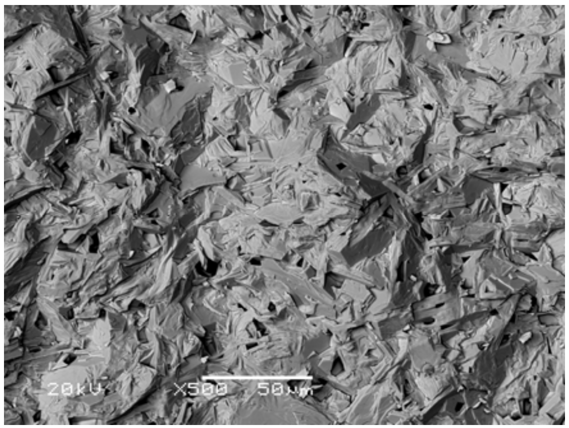
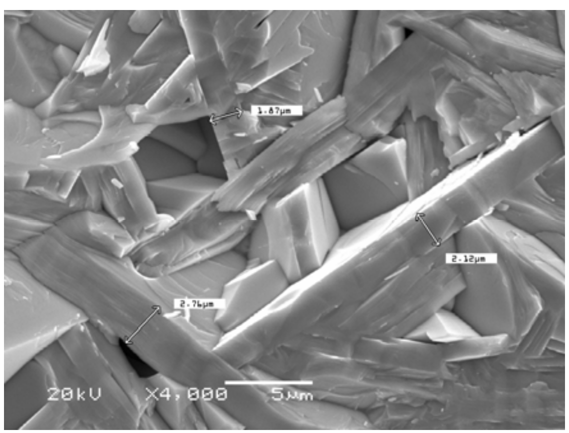
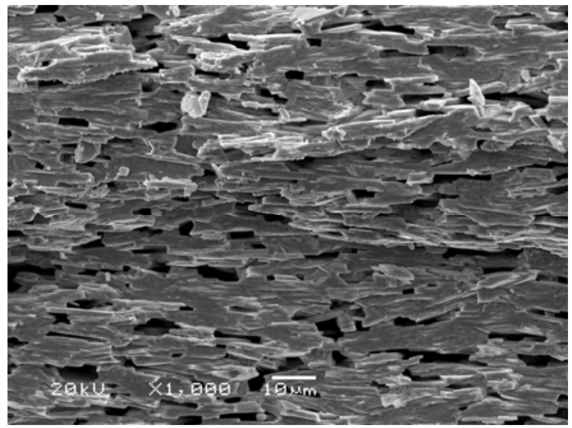
Progression from random to textured dense polycrystalline ceramic microstructures

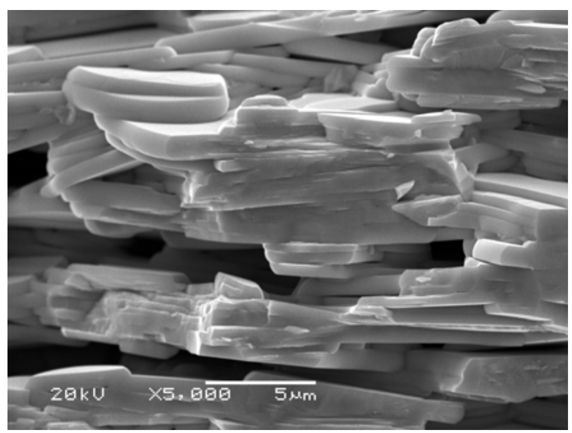
- Random polycrystalline dense microstructure



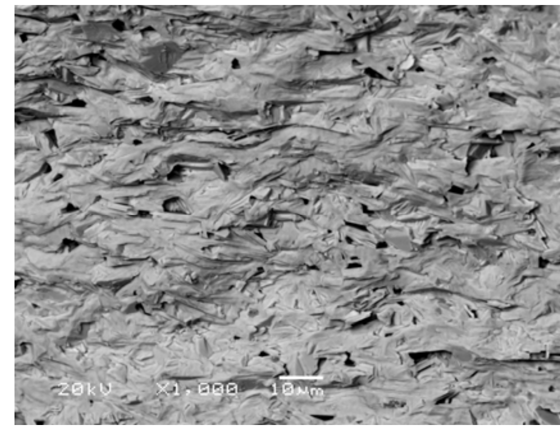


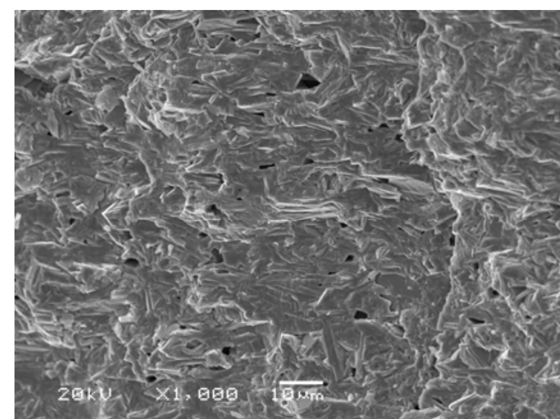
- Textured polycrystalline low-density microstructure





- Textured polycrystalline dense microstructure

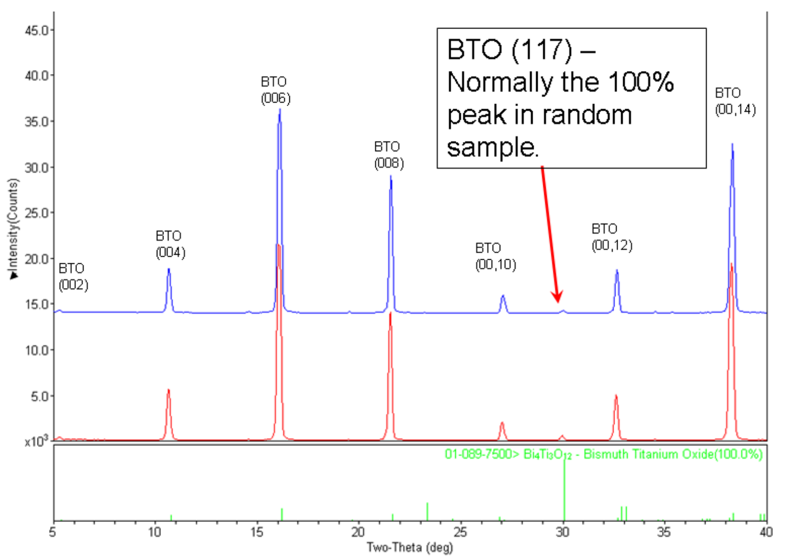


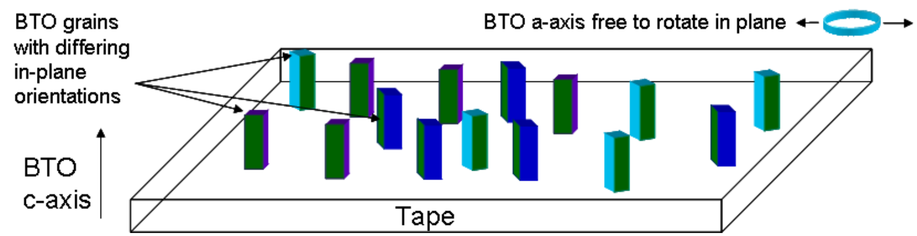


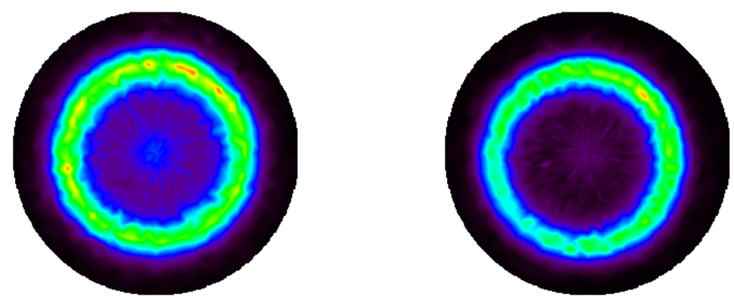
Laboratories

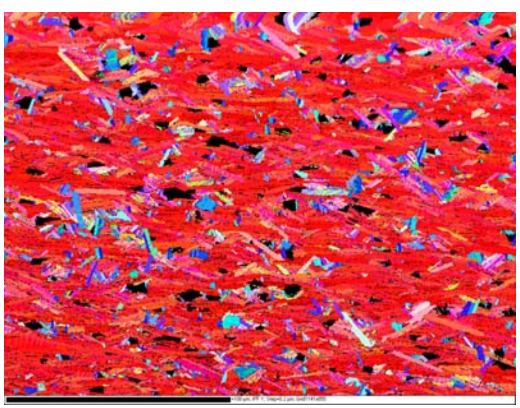
Confirmation of the degree of textured microstructure

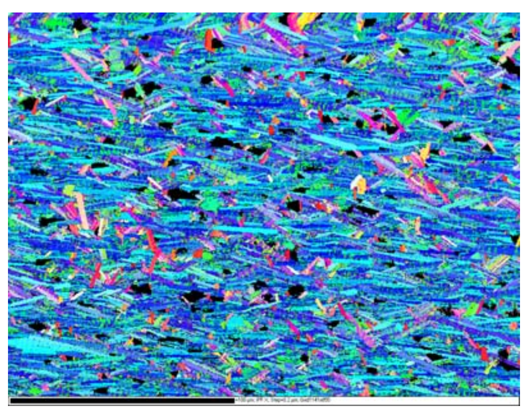
- Texturing Results evaluated through XRD (rocking curves, chi-tilt, pole figures) and EBSD
  - Tape casting – 0.70-0.95 based on Lotgering factor with sintered densities between 90-98% TD
  - Screen printing – 0.90-0.96 based on Lotgering factor with film quality and density TBD

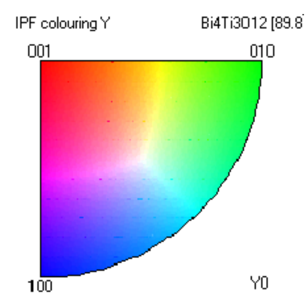


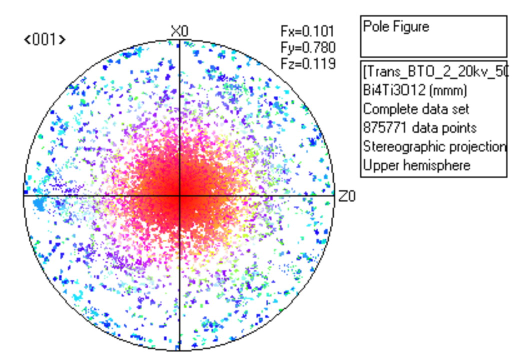








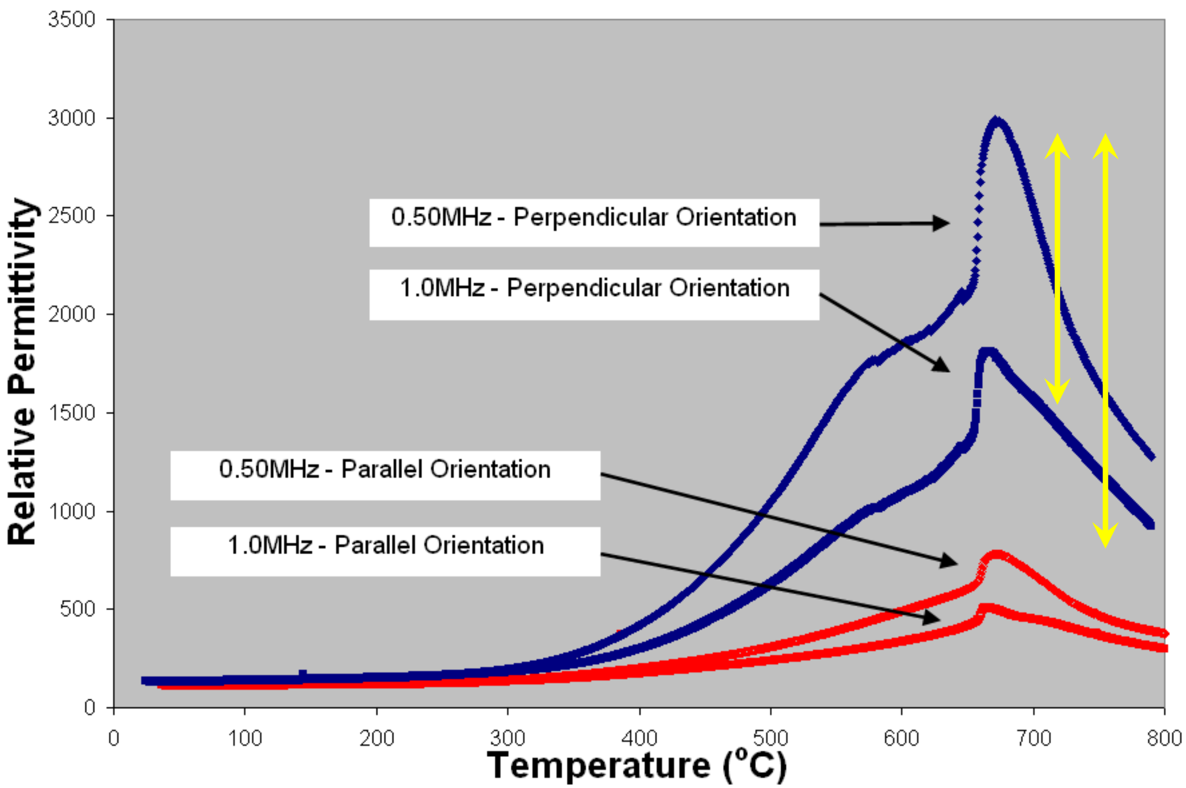




Anisotropic weak-field dielectric behavior

- Weak-Field Dielectric Properties – BTO System

Temperature (°C)	Frequency (MHz)	Orientation	Relative Permittivity	% Difference
670	0.5	Parallel	778	
670	1	Parallel	503	
670	0.5	Perpendicular	2980	73.89%
670	1	Perpendicular	1803	72.10%
670	0.5	Random Polycrystalline	1168	60.81%
670	1	Random Polycrystalline	849	52.91%

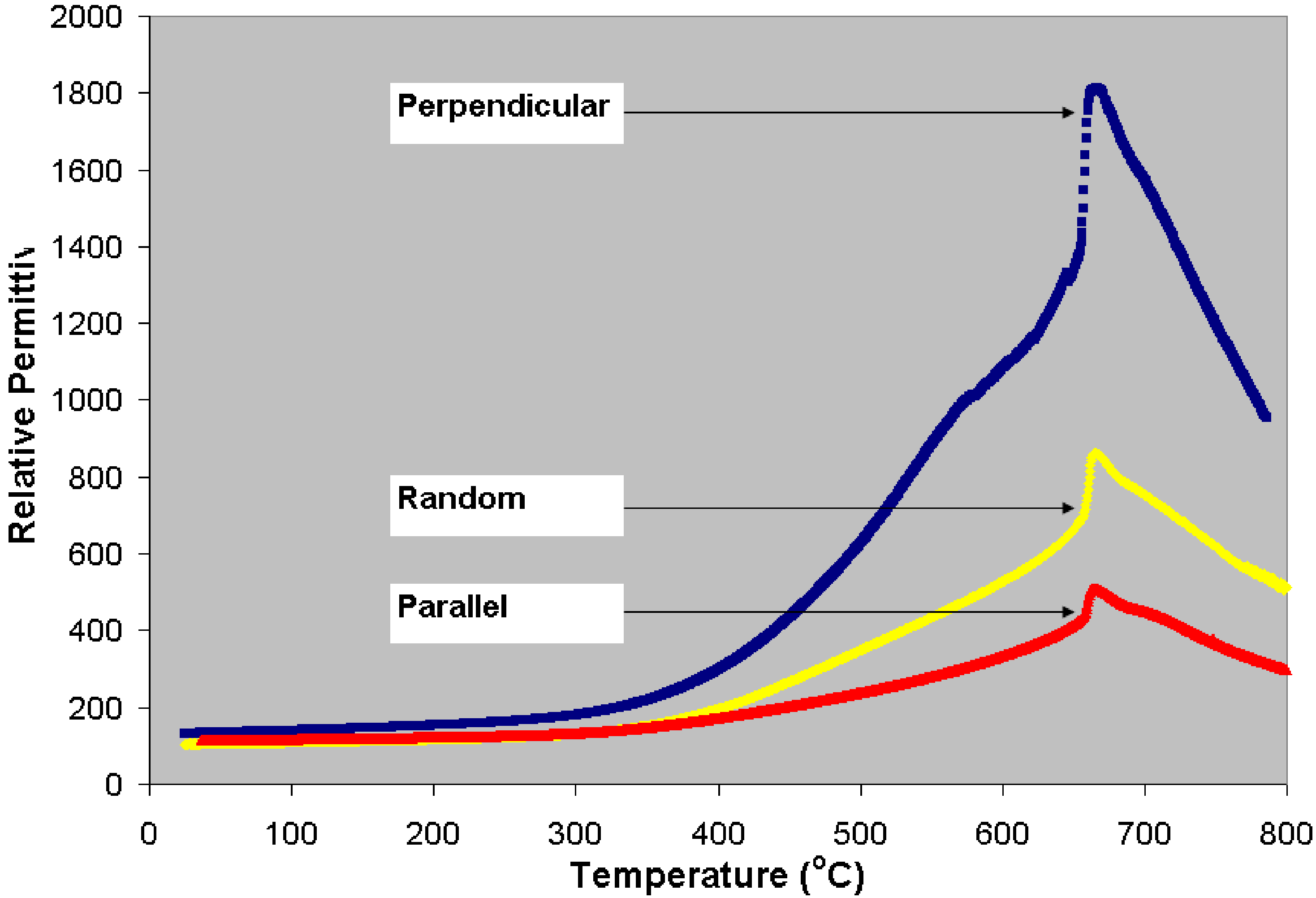


≈73% difference observed Perpendicular-Parallel Orientation

≈60% difference observed Random Polycrystalline-Perpendicular Orientation

Anisotropic weak-field dielectric behavior

- Weak-Field Dielectric Properties – BTO System



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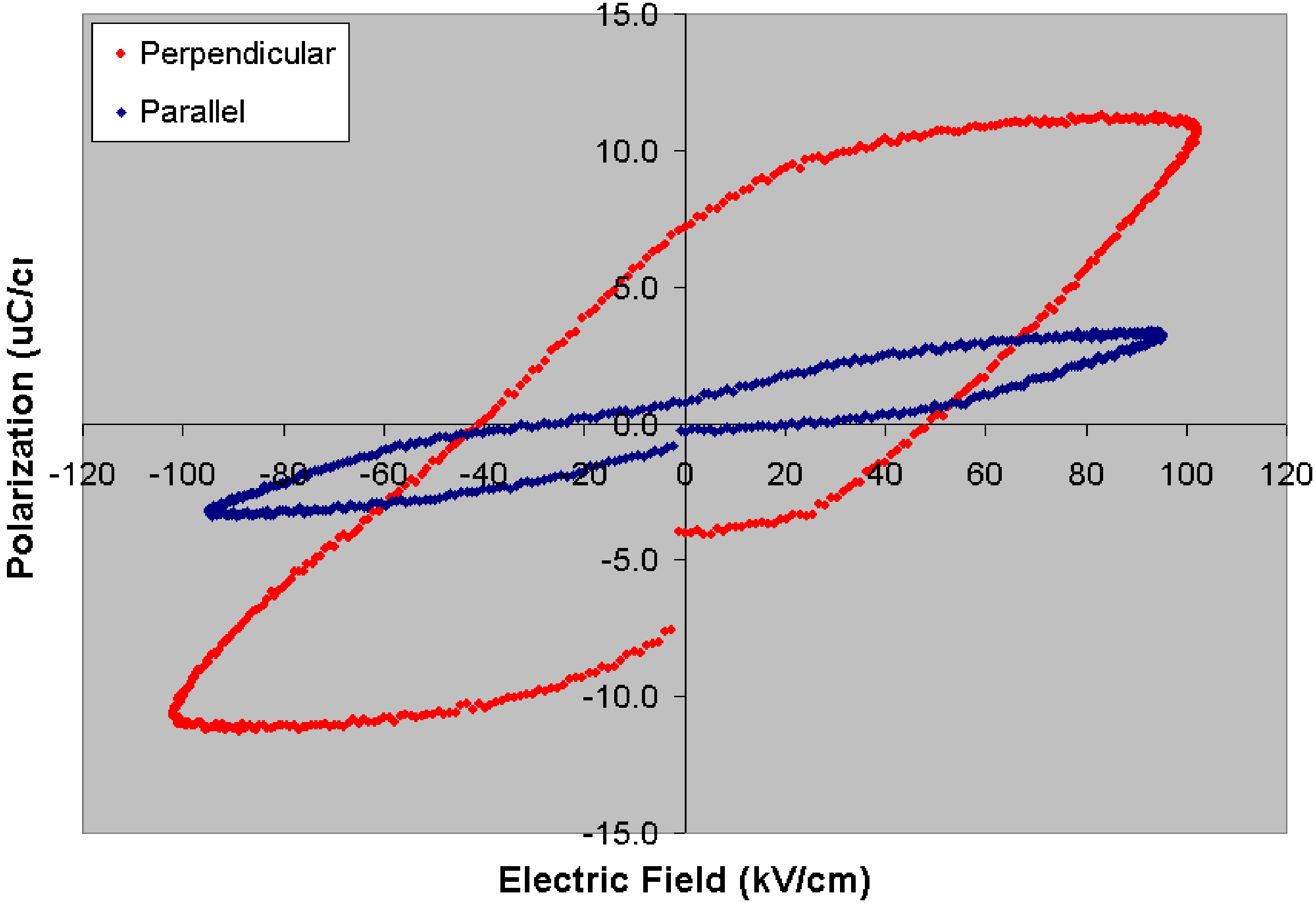
Anisotropic polarization behavior as a function of applied electric field

- Polarization as a function of applied electric field
  - Frequency = 1 Hz, room temperature



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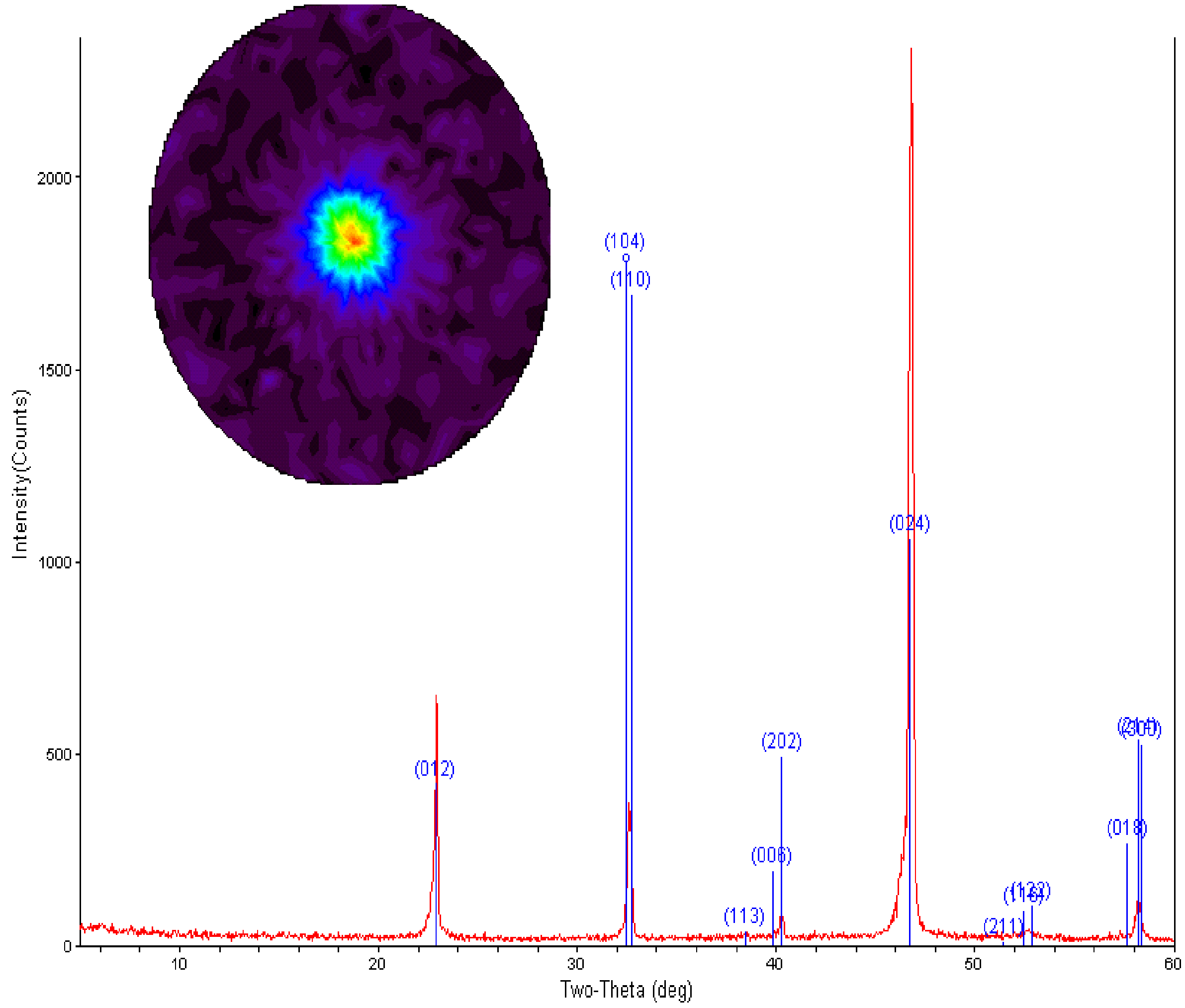
X-ray Diffraction θ-2θ scan and pole figure analysis for templated NBT

- 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^o 2θ



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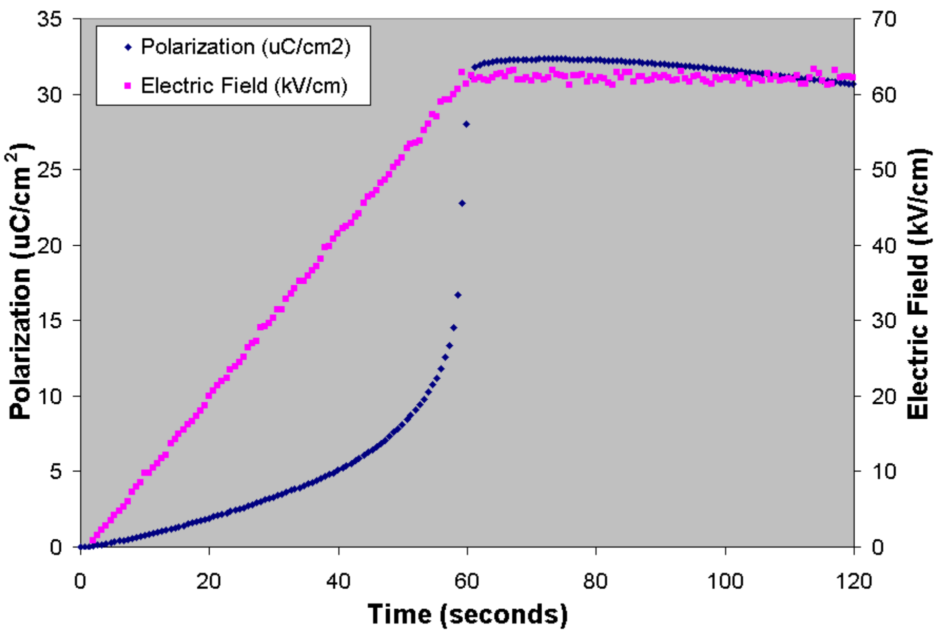
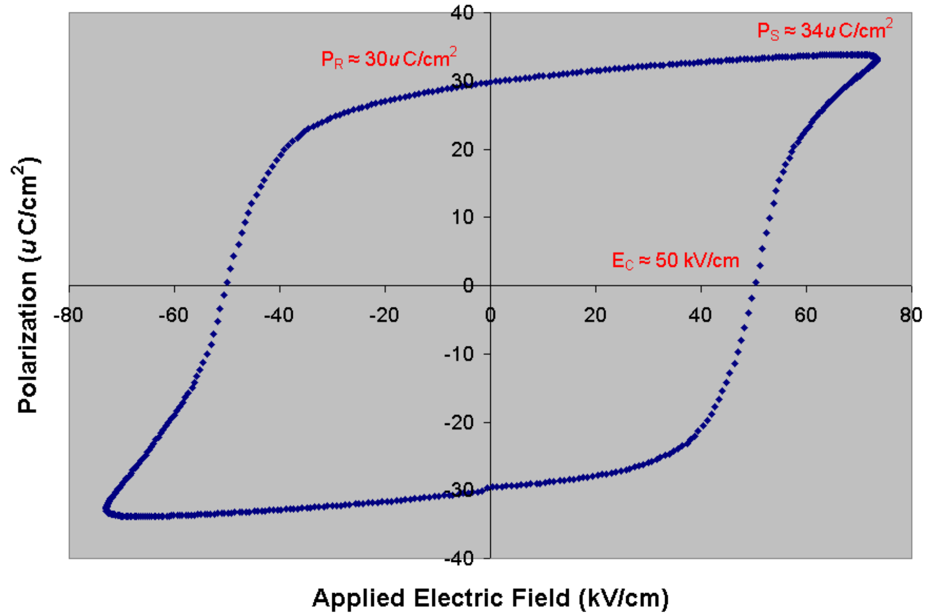
High Field Poling and Polarization Behavior of NBT

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





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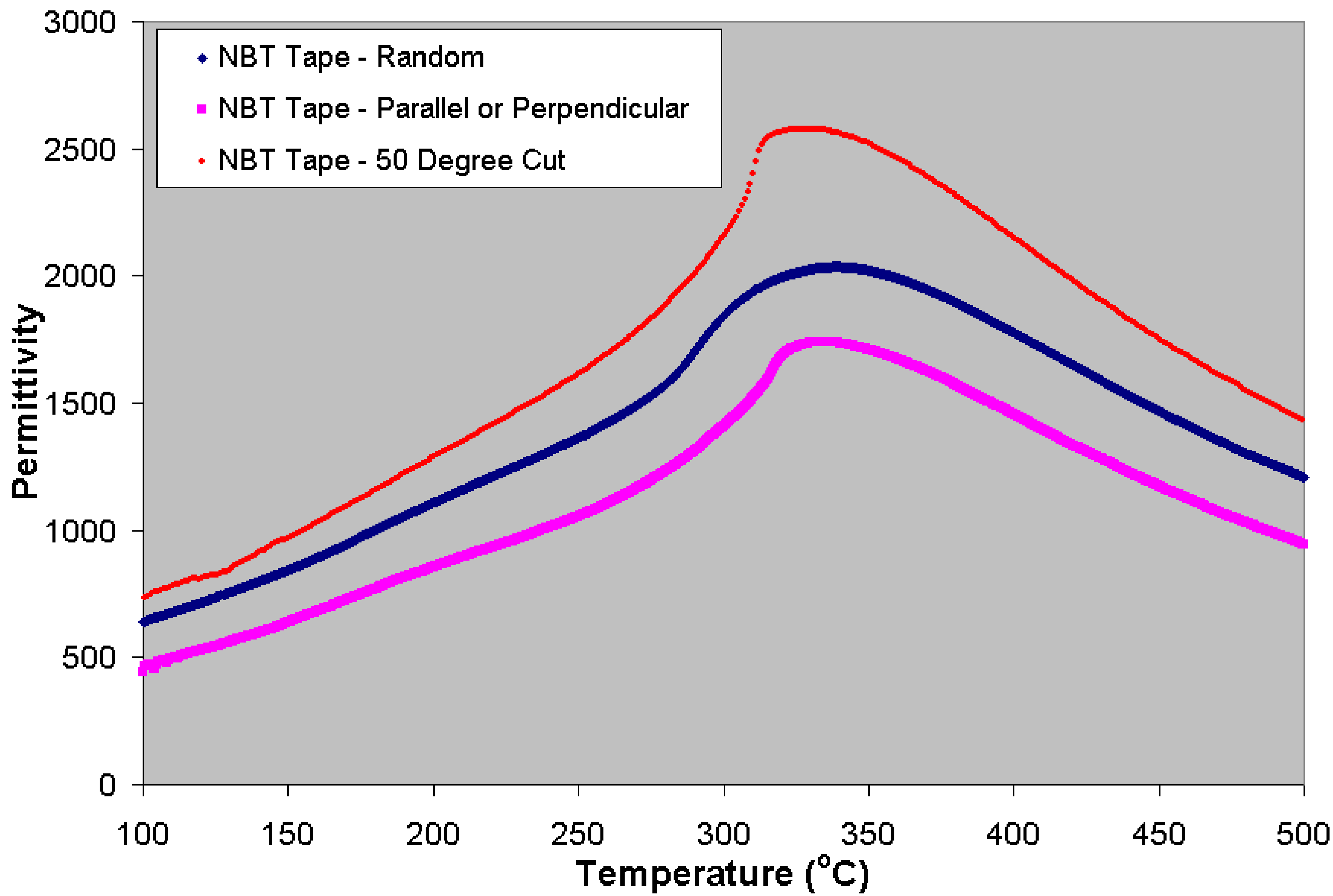
Anisotropic weak-field dielectric behavior

- Weak-Field Dielectric Properties – NBT System



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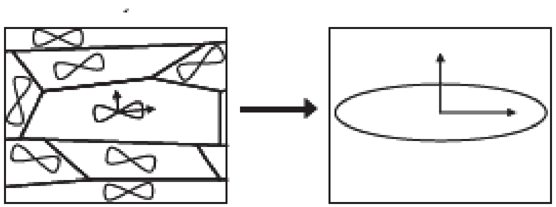
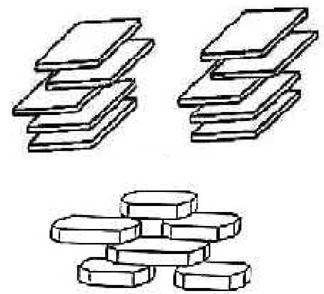
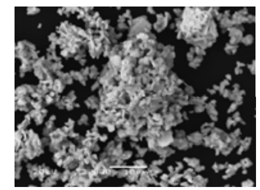
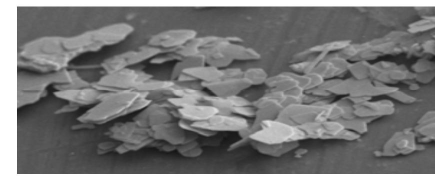
Microstructural engineering through processing induced texturing

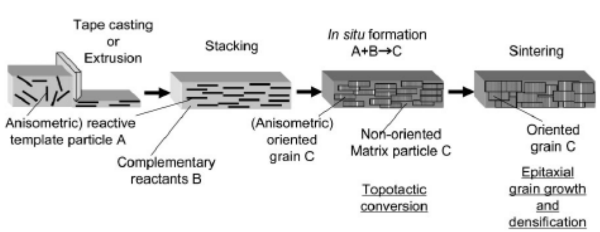
- Target – Develop an advanced, low cost manufacturing process for a non-lead based microstructural engineered ferroelectric ceramic material
  - Microstructural engineering through texture modifications

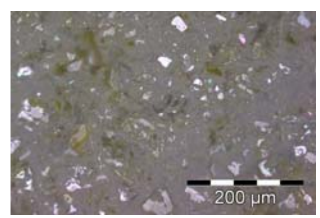


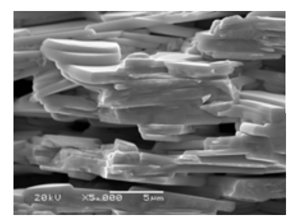


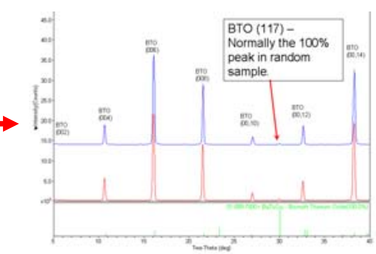


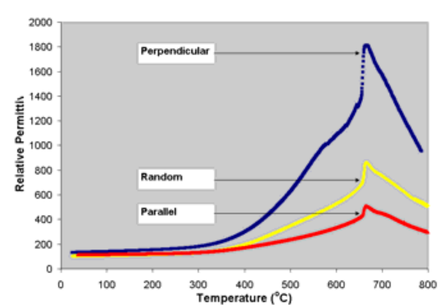












J.L. Jones, "Texture and Anisotropy of Polycrystalline Piezoelectrics", JACERS, 90 [8] 2297-2314 (2007)

Enhanced Ferroelectric and Dielectric Property Relationships Induced by Textured Processing for Several Ceramic Compositions

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