Creating High Temperature Ferroelectrics by Using Ternary Components with Limited Solubility

Benjamin Hirt[1], Benjamin Kowalski[2], Alp Sehirlioglu[1]

[1]US Air Force/Case Western Reserve University, 10900 Euclid Ave, Cleveland, 44106 USA [2]NASA Glenn Research Center, 21000 Brookpark Rd., Cleveland, OH, 44135 USA

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Motivation

- High temperature ferroelectrics are needed for actuator applications ranging from drilling on the surface of Venus to fuel modulation in jet engines
- Machine learning in previous work has dramatically reduced search space and produced new candidates for experimentation.

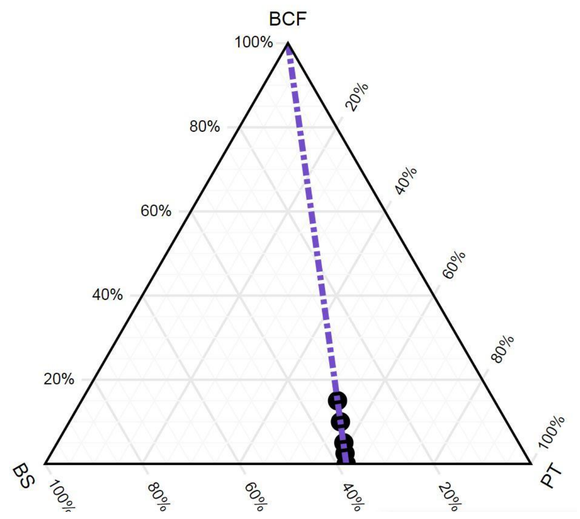
Objective

Create high temperature piezoelectrics by changing end member of ternary systems. Starting point was near MPB between $BiScO_3$(BS) and $PbTiO_3$ (PT) (38%/62%). Third components suggested by machine learning to create a high T_c with limited solubility; $BiCo_{0.88}Fe_{0.12}O_3$ (BCF)[1]

Experiment

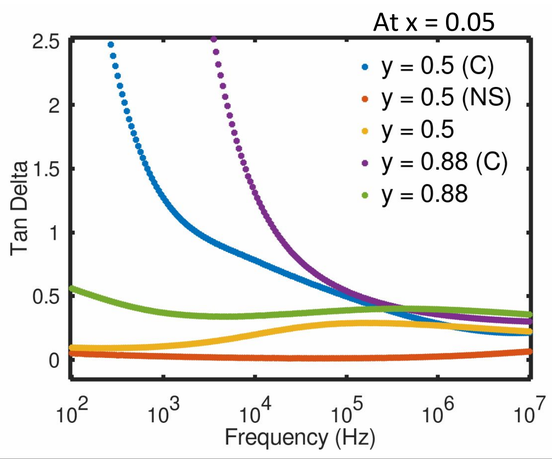
$$(x)(0.38BiScO_3 - 0.62PbTiO_3) - (1\text{-}x)Bi(Co_yFe_{1\text{-}y})O_3$$

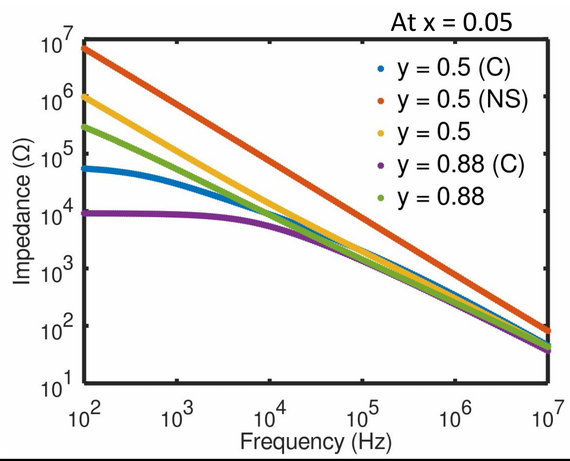
- x was varied between 0.00 and 0.15; dense samples were not attained at x = 0.15
- y was varied from 0.88 to 0.50 to lower both ac and dc conductivity
- Samples were prepared by conventional power processing. Calcination at 750°C for 3 hours. Sintering at 1100°C for 1 hour



The Cobalt Problem

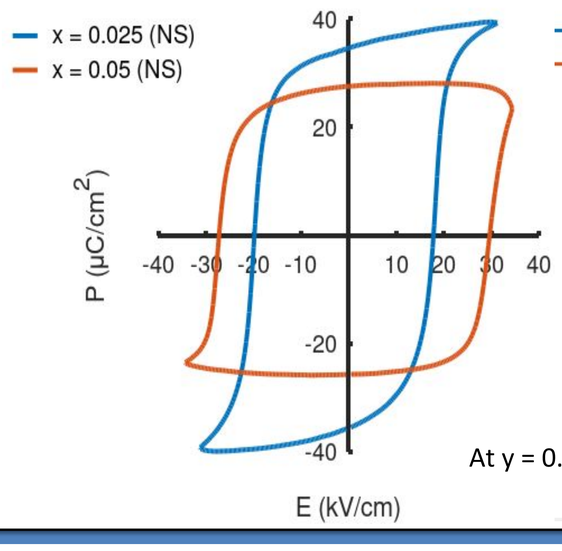
- At high Co content (y = 0.88) samples were too conductive to pole. Therefore, Co content was reduced to y = 0.50
- To compensate for Co volatility +5% mol Co (C) was added
- A Pb/Bi containing secondary phase was identified. Non-stoichiometric (NS) compositions were prepared by removing -5% mol Bi/Pb

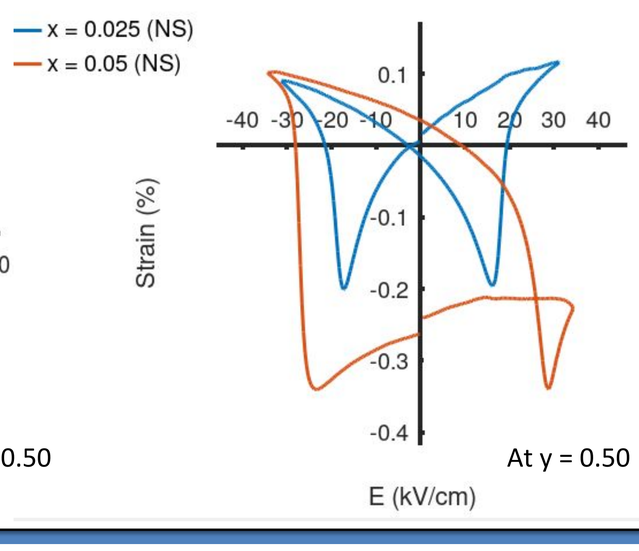




Pinning of domains with increased BCF content

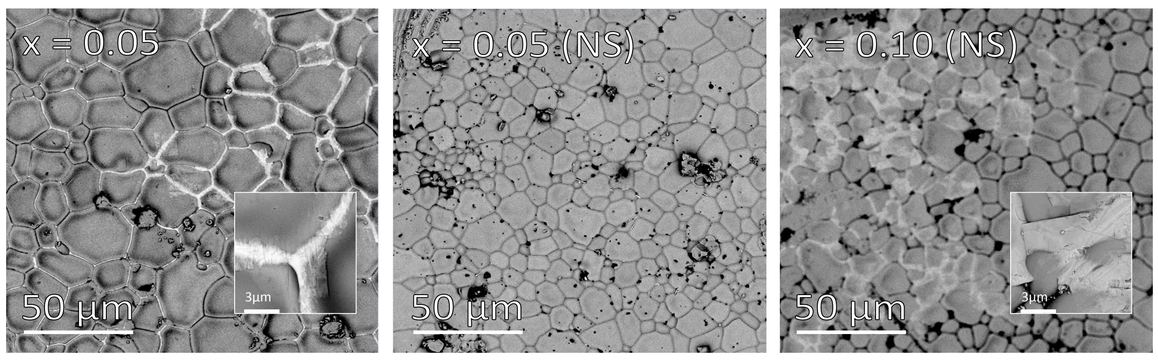
- Ternary component has a pinning effect, making it harder to pole
- Switches from Rhombohedral (R) to Tetragonal (T) as x increases
  - 100% R at x = 0 / 47% R at x = 0.05 / 29% at x = 0.10 / 0% at x = 0.15
- 5% mol removal of Bi/Pb helps reduce T content
  - 63% R at x = 0.05 (NS) / 47% R at x = 0.05
- Decrease in y also helps lower T content
  - at x = 0.05: 28% R at y = 0.88 / 47% R at y = 0.50
  - at y = 0.50: 100% R at x = 0.025 (NS)



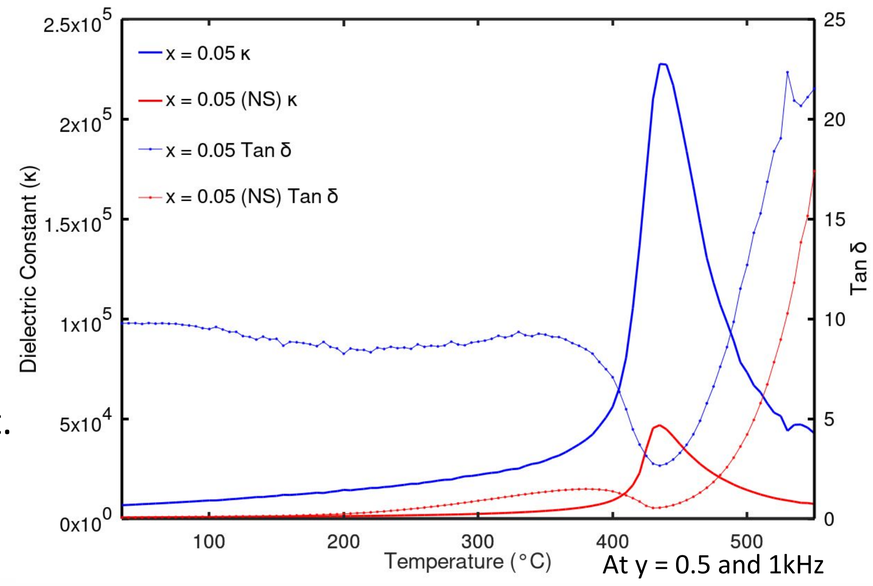


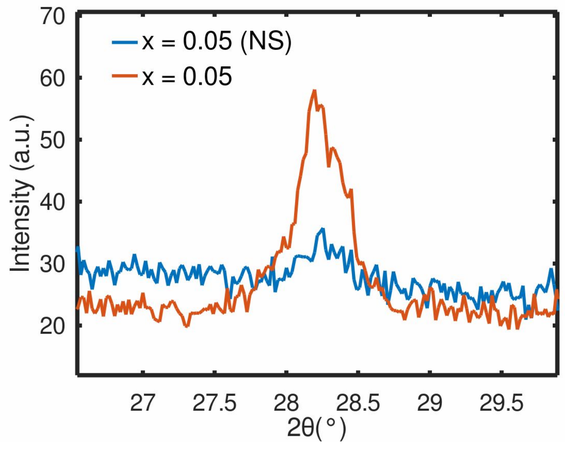
Secondary Phase Problems

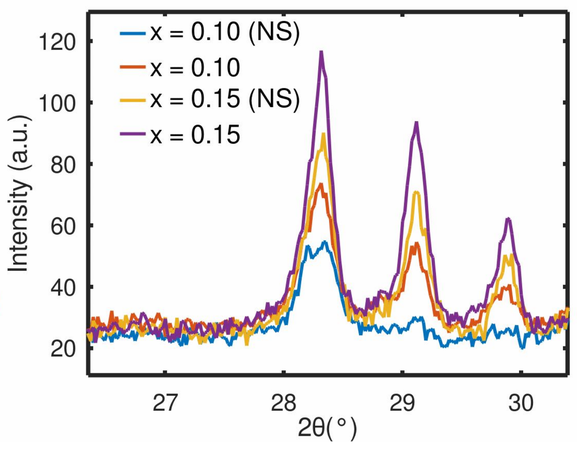
- Secondary phase containing Bi/Pb forms between grain boundaries
- Secondary phase is conductive and continuous → makes poling impossible if there is more than ~5%



- Reduced secondary phase by removing 5% mol Bi and Pb
- Increase in impedance and decrease in loss tangent. Samples can be poled



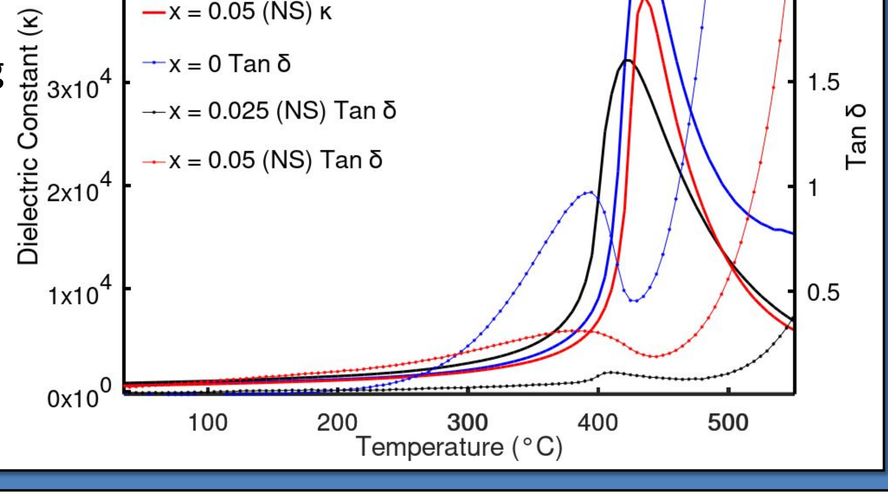




- Up to x = 0.05 (NS) the secondary phase is eliminated and can be poled. At higher x, secondary phase still occurs even with NS
- Without the removal of the second phase samples cannot be poled for ferroelectric measurements.

Compared to the Binary

- The loss tangent is much lower overall and especially at higher operating temperatures
- Slight decrease in T_c
- Overall allows increase in operating temperature



Conclusion

- Cobalt and parasitic secondary phases were causing poling problems
- Loss tangent was significantly decreased at higher temperatures, T_c did not increase, and was slightly reduced for some x

Reference

1.Balachandran, P.V., Kowalski, B., Sehirlioglu, A. et al. Experimental search for high-temperature ferroelectric perovskites guided by two-step machine learning. Nat Commun 9, 1668 (2018) doi:10.1038/s41467-018-03821-9.

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