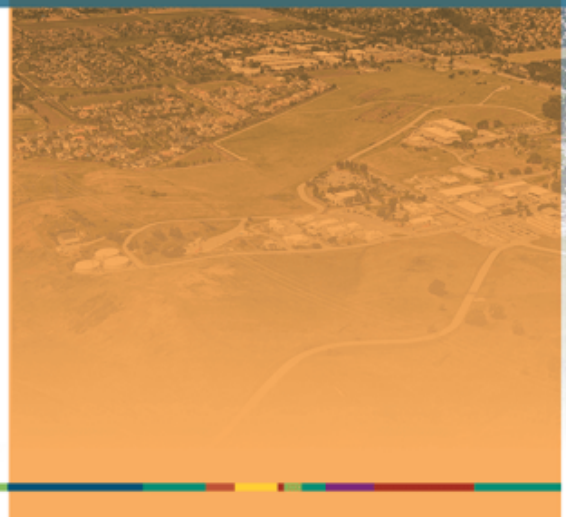
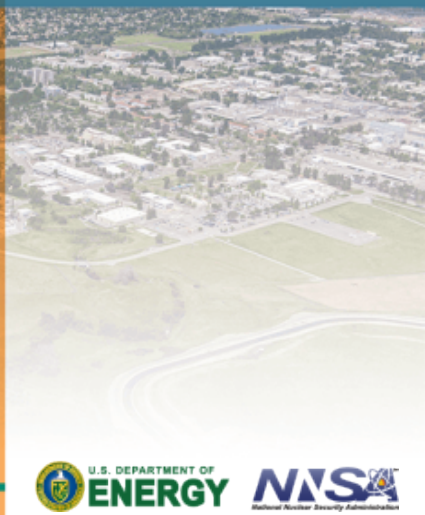
SAND2022-0249C

Sandia National Laboratories

DC lifetime and thermally stimulated depolarization current (TSDC) of $Bi(Zn,Ti)O_3$-$BaTiO_3$ (BZT-BT)





Sean Bishop, Mia Blea-Kirby, Amanda Peretti, Luis Jauregui, Will Bachman, and Jon Bock

Electronic Materials and Applications 2022 (EMA 2022) - ACerS

January 20, 2022
Room Citrus A / Pre-Recorded
4:15 - 4:30 PM

U.S. DEPARTMENT OF ENERGY | NNSA

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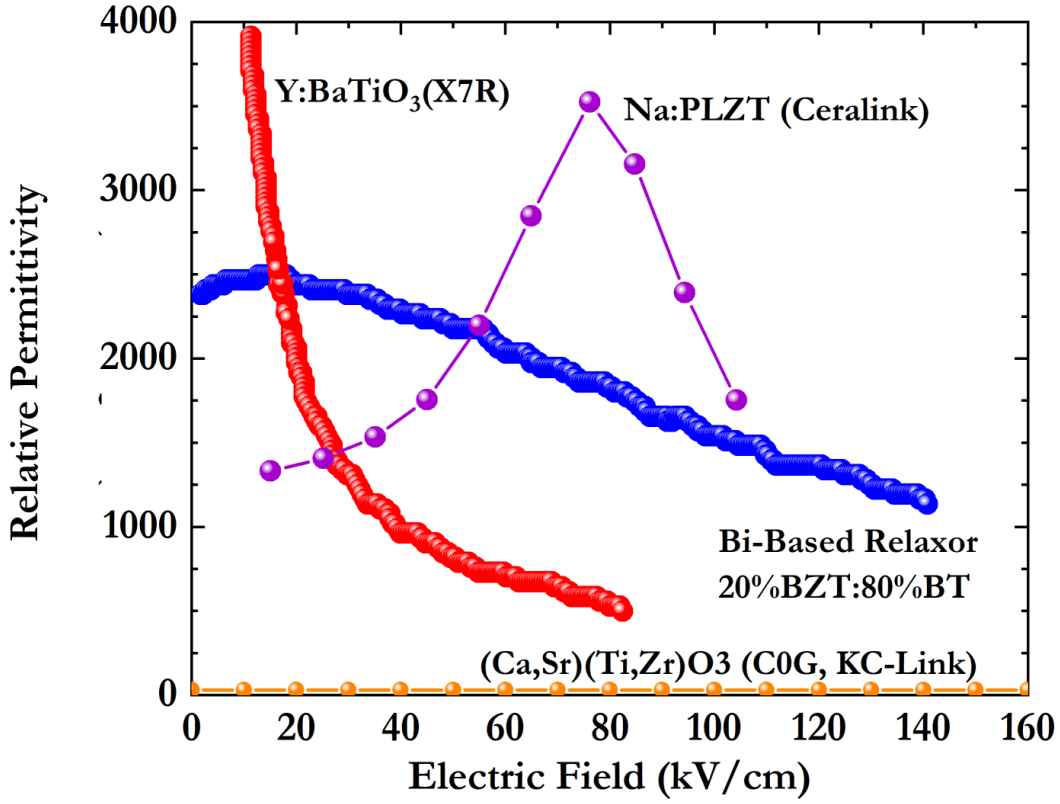
High Permittivity in High Electric Field and Temperature

20BZT-BT: 20% $Bi(Zn_{0.5}Ti_{0.5})O_3$-80% $BaTiO_3$

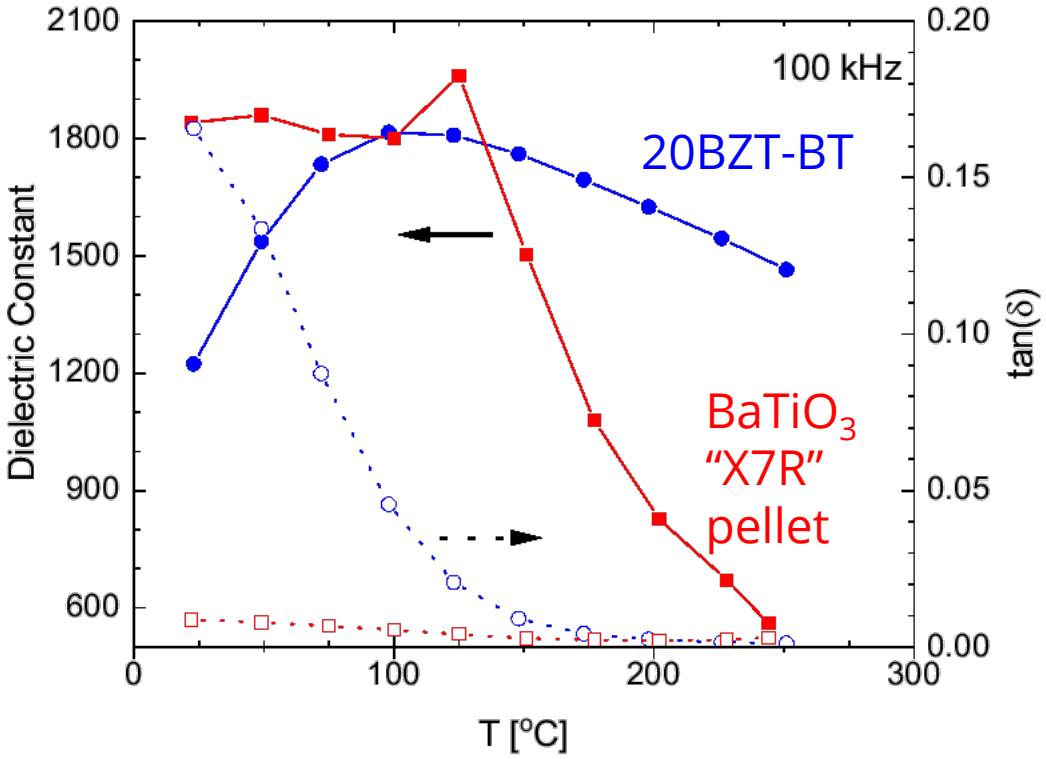
High and less variable permittivity at high electric field and temperature, desirable for high power conversion applications

High permittivity at >120 kV/cm



J. Am. Ceram. Soc., 99 (2016) 2849

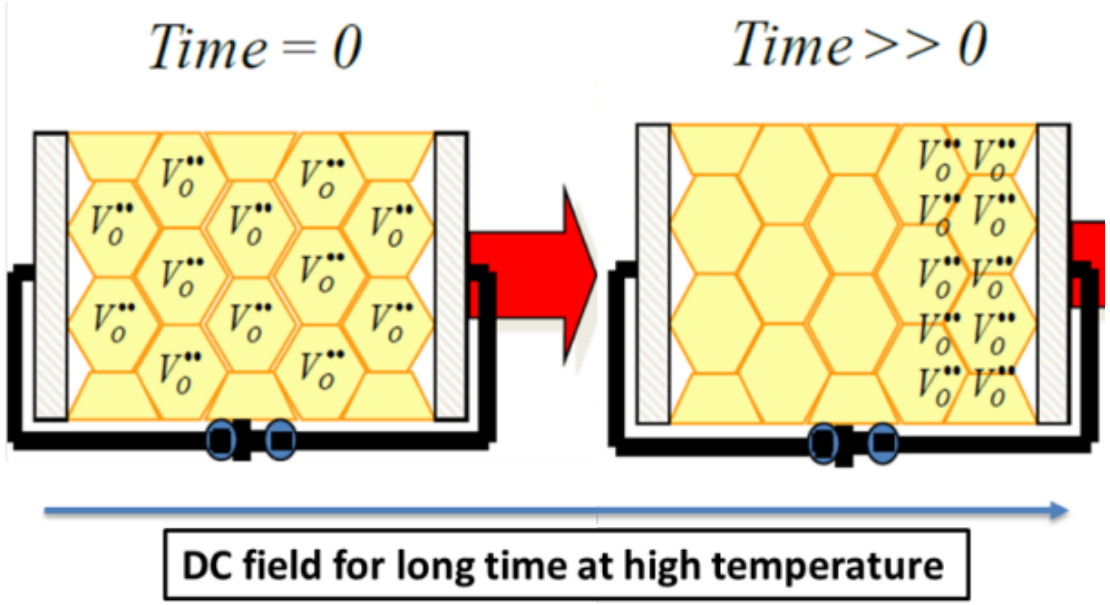
High permittivity at >200°C at 100 kHz



Oxygen Vacancy Migration Can Limit DC Lifetime

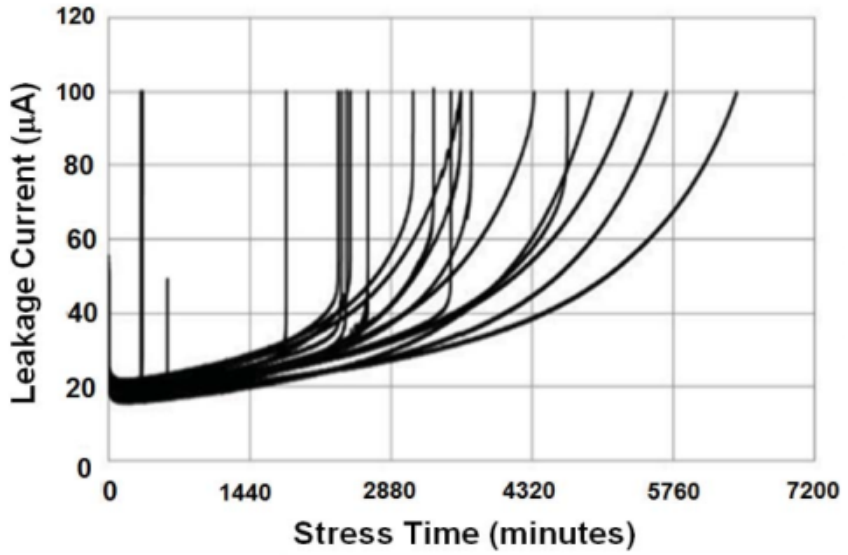
DC bias → oxygen vacancy (V_O^{**}) migration

- Change in electron and hole concentration
- Increase in bulk and or electrode leakage current



R. Maier, Penn. State PhD Thesis (2014)

Accelerated Lifetime for BME X7R MLCC's



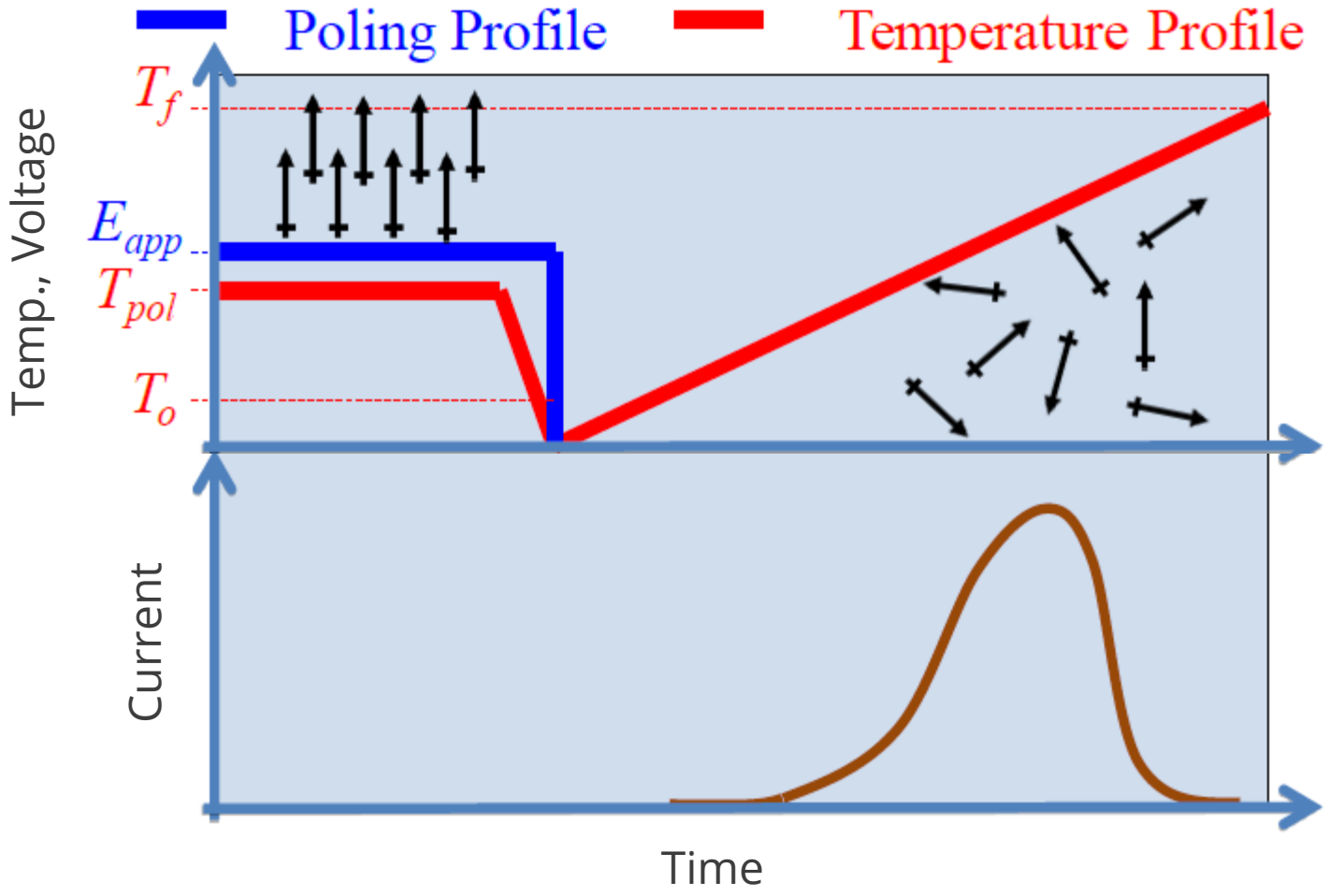
Accelerated test (5x Vr)
0.47 uF, 50 V Rated, 0805, 250 V, 155 °C

D. Liu, IEEE Tran. Comp. Pack. Man. Tech. 5 (2015) 40

DC lifetime and degradation modes uncharacterized in BZT-BT

Thermally Stimulated Depolarization Current (TSDC)

V_O^{**} are typically not majority charge carriers → probe mobile point defects with TSDC

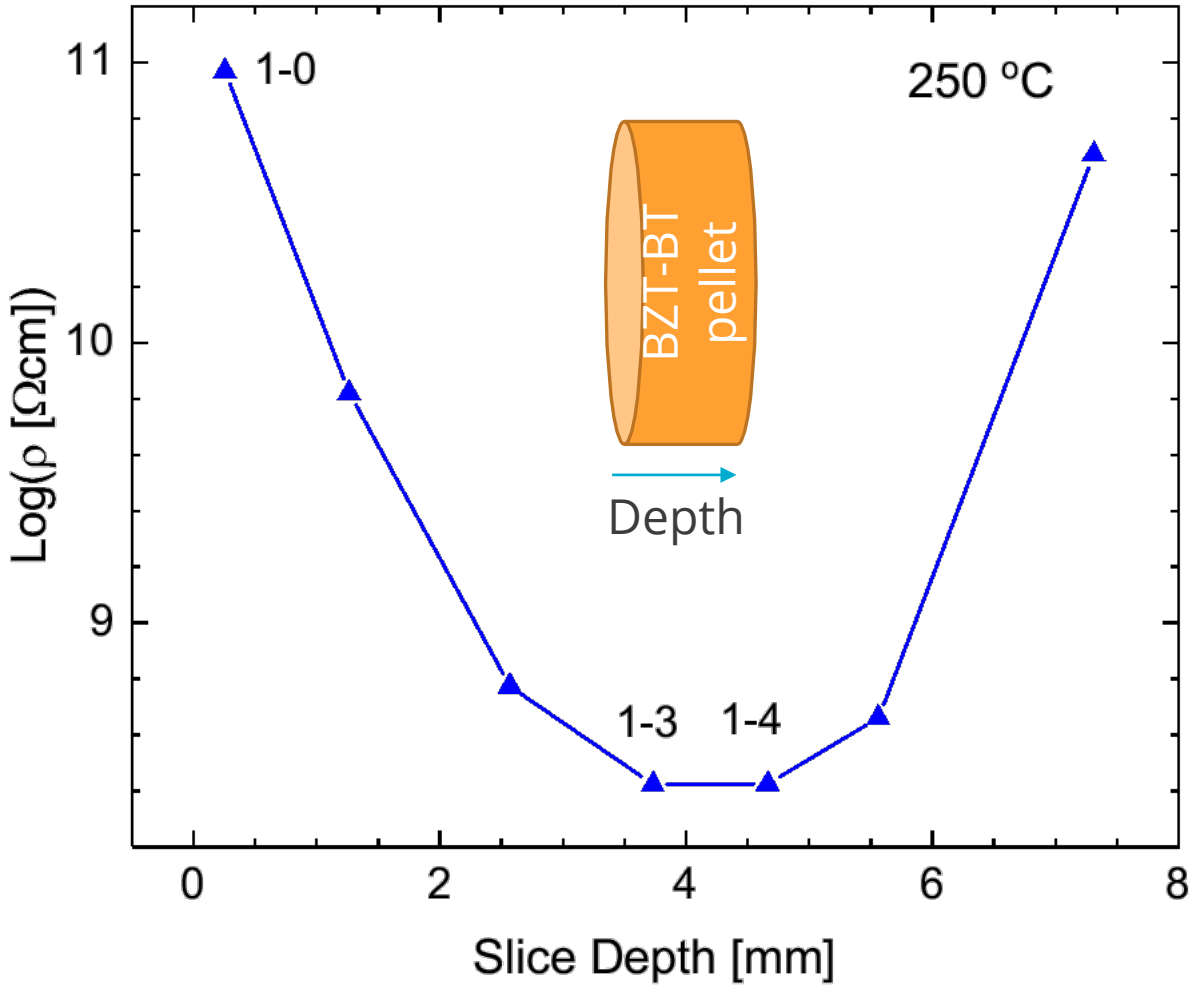


R. Maier, Penn. State PhD Thesis (2014)

BZT-BT

Bi-rich BZT-BT: $(Bi_{0.22}Ba_{0.78})(Zn_{0.1}Ti_{0.9})O_3$

Donor doped to suppress oxygen vacancy concentration

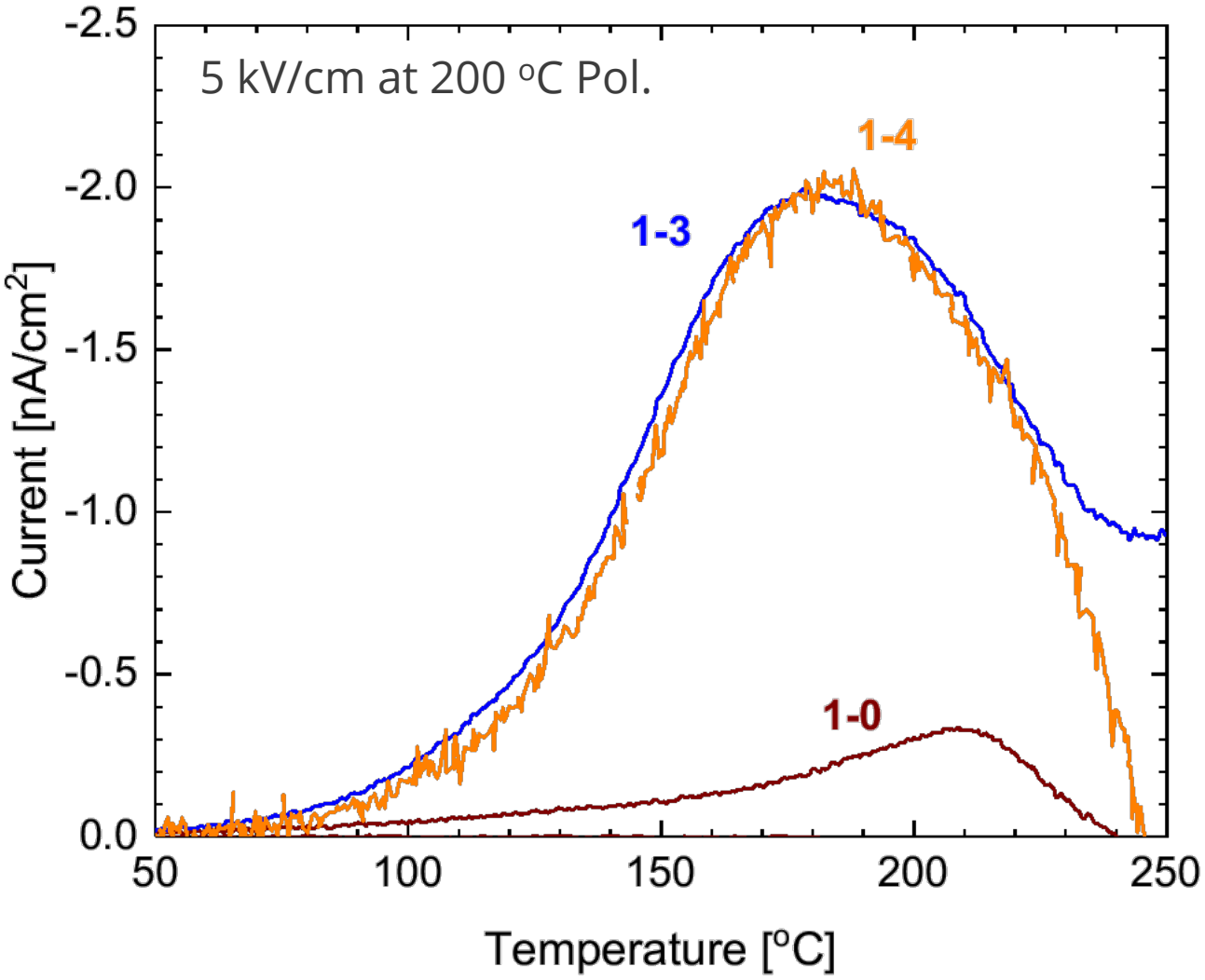


- Significant resistivity decrease in center of thick sample
- Likely reduced in center from frozen in V_O^{**} from high temperature sintering or residual organic binder

$$O_O^X \rightarrow V_O^{**} + 2e^- + \tfrac{1}{2}O_2\,(g)$$

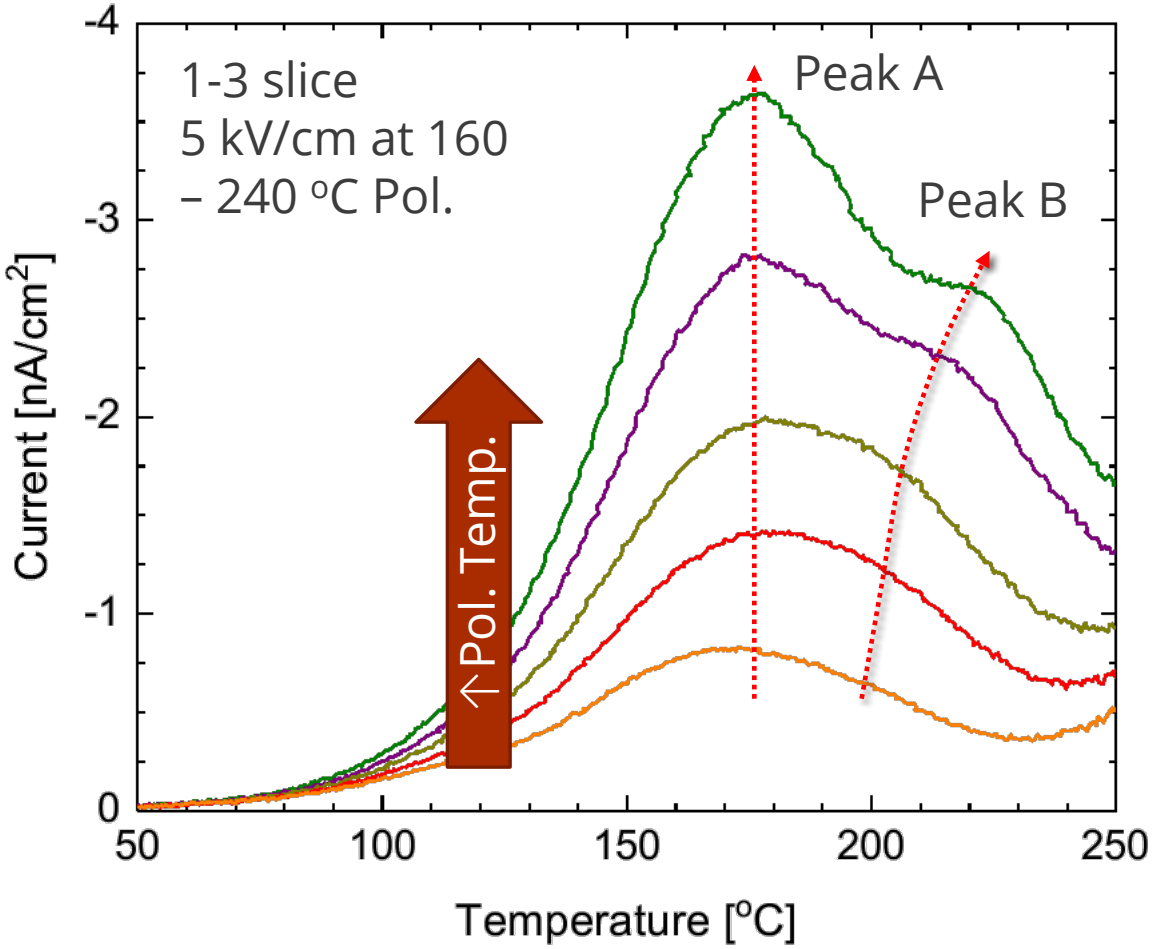
→ Opportunity to examine V_O^{**} impact on TSDC and DC lifetime!

Largest TSDC Response Near Center of Pellet

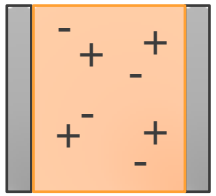


Very large TSDC near center of sample, consistent with significant V_O^{**} concentration

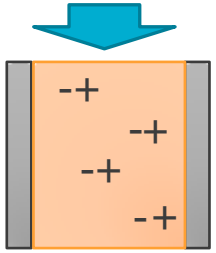
Polarization Temperature Dependence of TSDC



Dipole defects (e.g., V_O^{**}-V_{Ba}'')



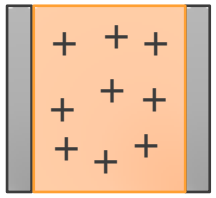
Polarize



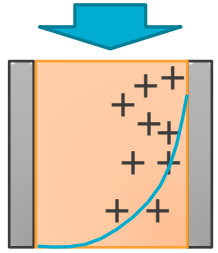
Defects "depolarize" at single temperature

Peak A

Space charge defects (e.g., V_O^{**})



Polarize



Greater concentration gradient → more time/temp. to depolarize

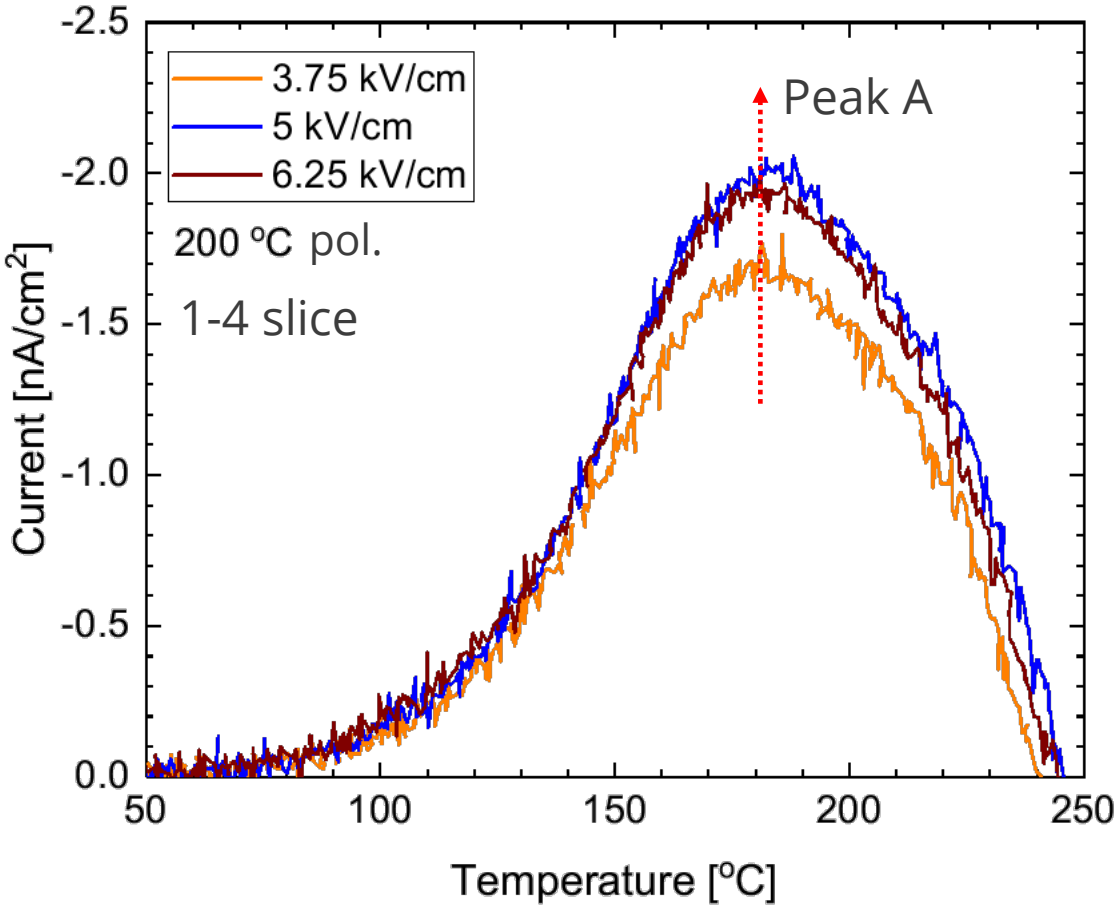
Peak B

- Limited temperature dependence of peak A suggests it is a dipole defect (e.g., trapped V_O^{**})[1]
- Increase in peak B with polarization temperature suggests it is from space charge[1]

Both peaks likely related to V_O^{**}

[1] *W.-E. Liu, PhD Thesis Penn. State (2009)*

Electric Field Dependence of TSDC

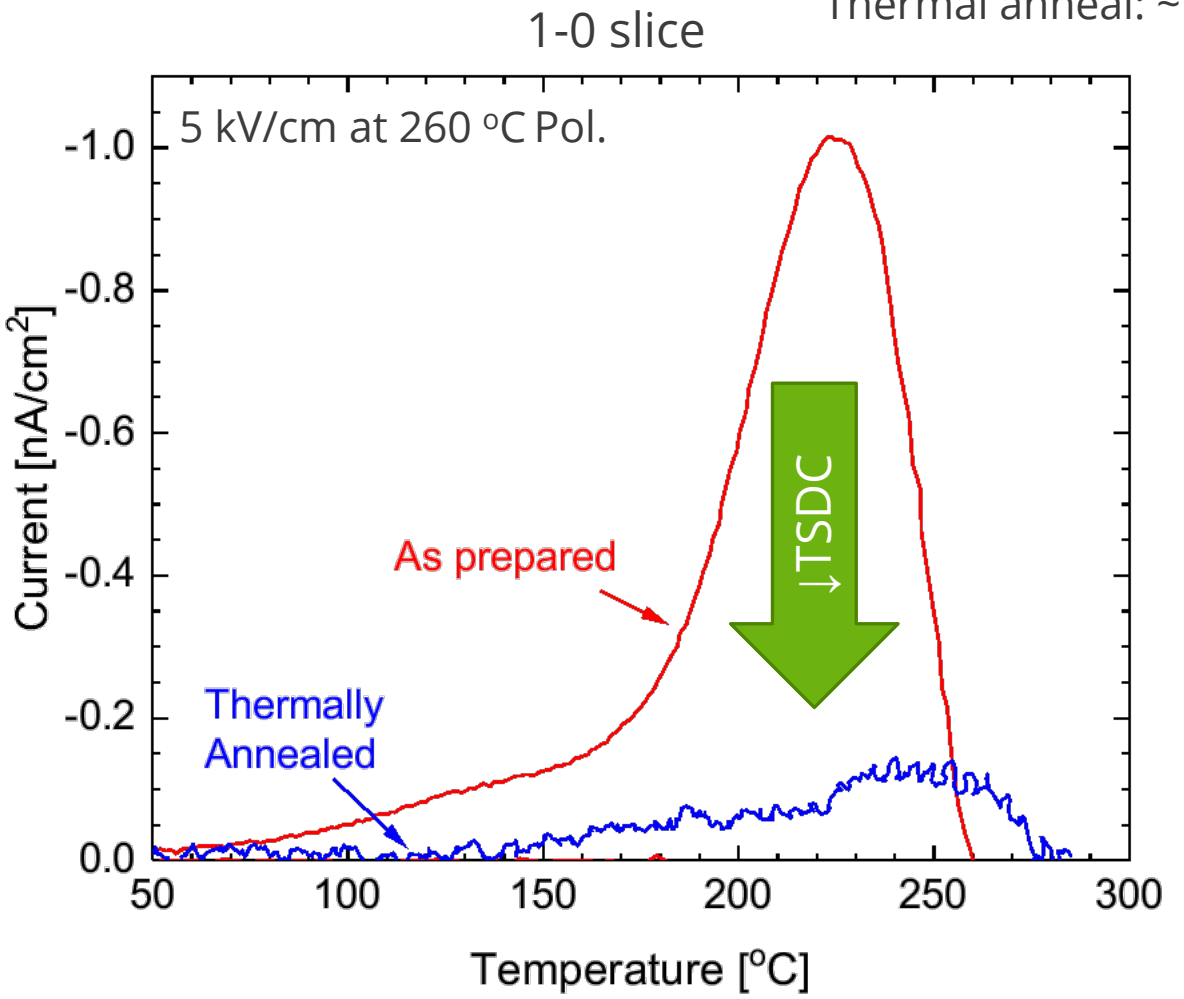


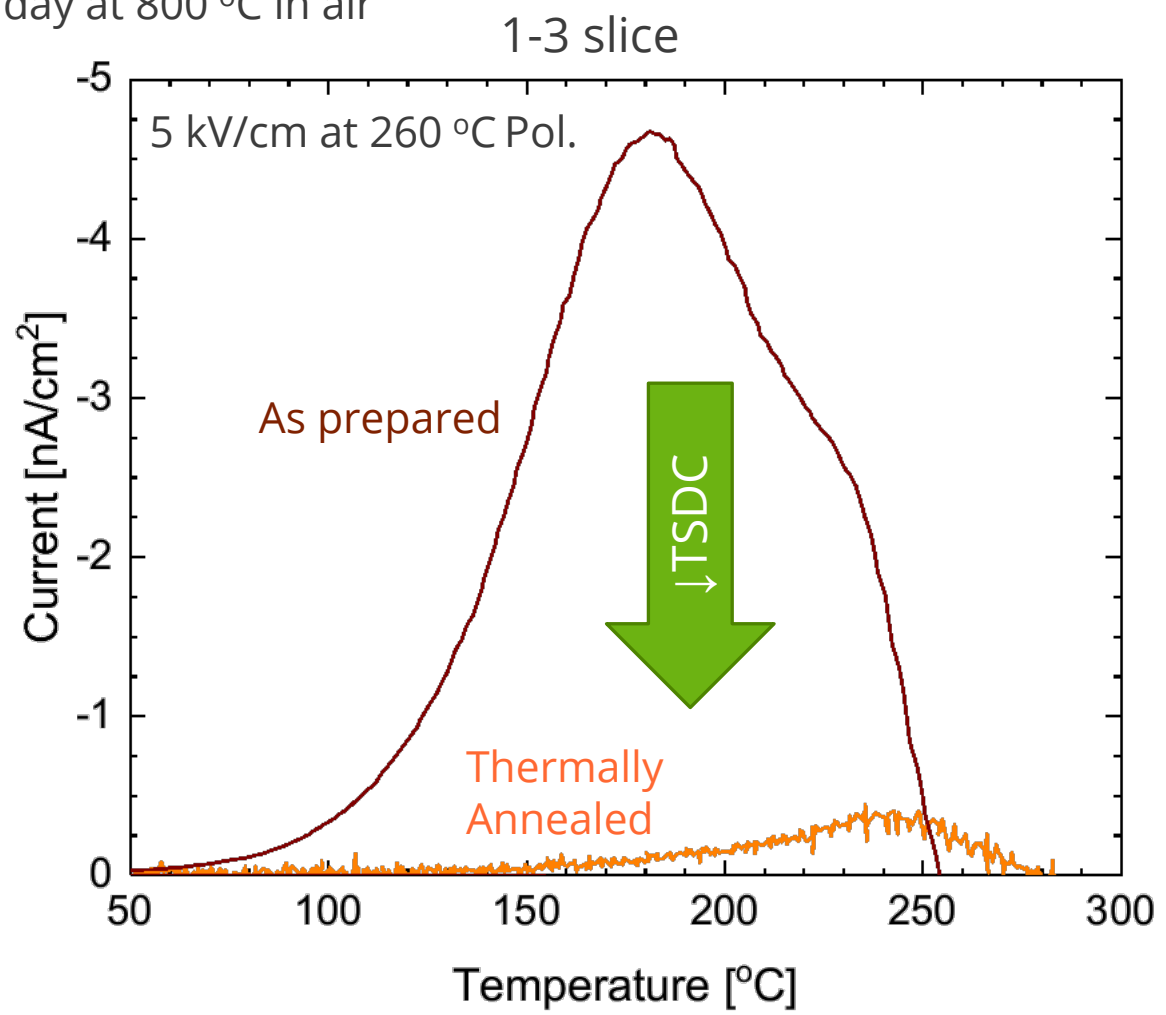
- Limited polarization field dependence of peak A indicates dipole defects[1]
- Apparent saturation in peak indicates critical field at which dipole defects separate

[1]*W.-E. Liu, PhD Thesis Penn. State (2009)*

Impact of Oxidative Thermal Annealing on TSDC

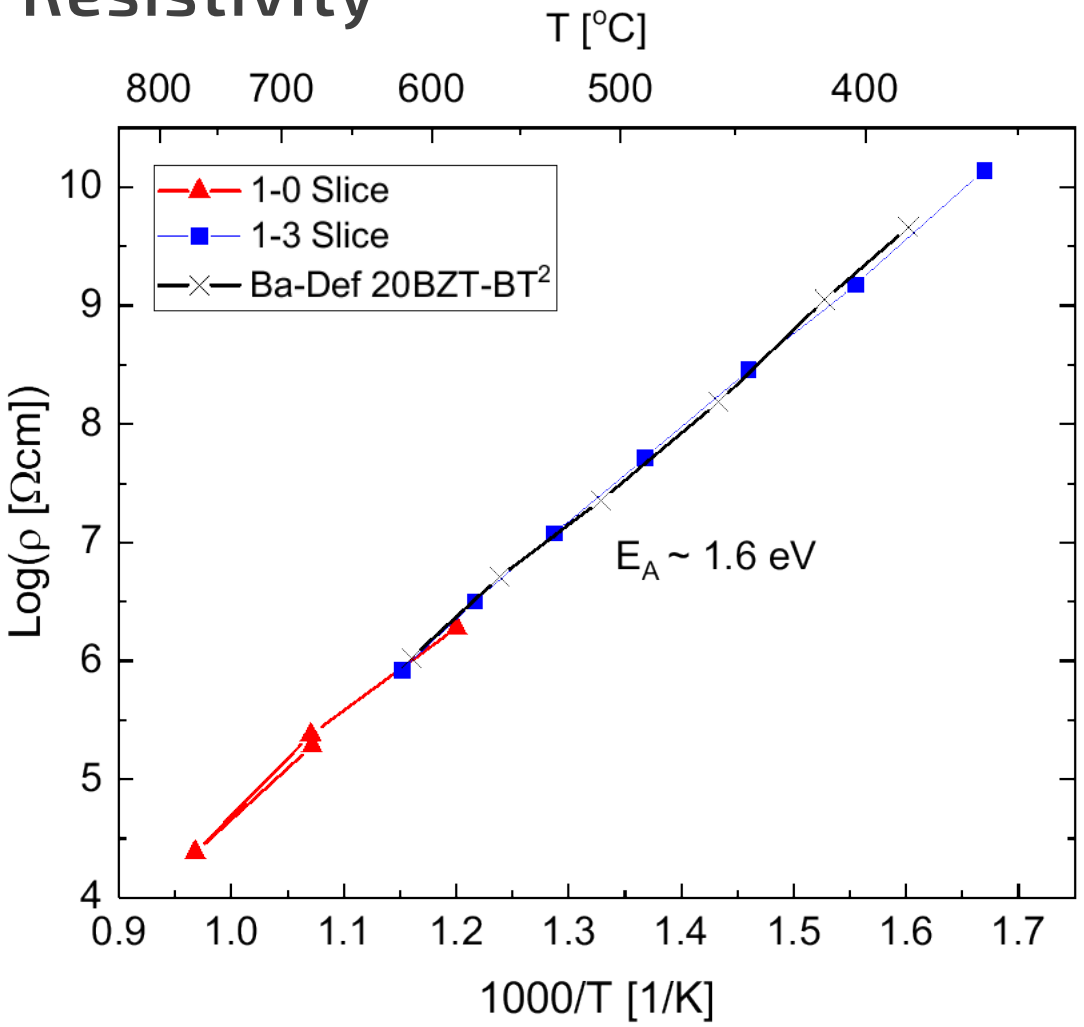
Thermal anneal: ~1 day at 800 °C in air

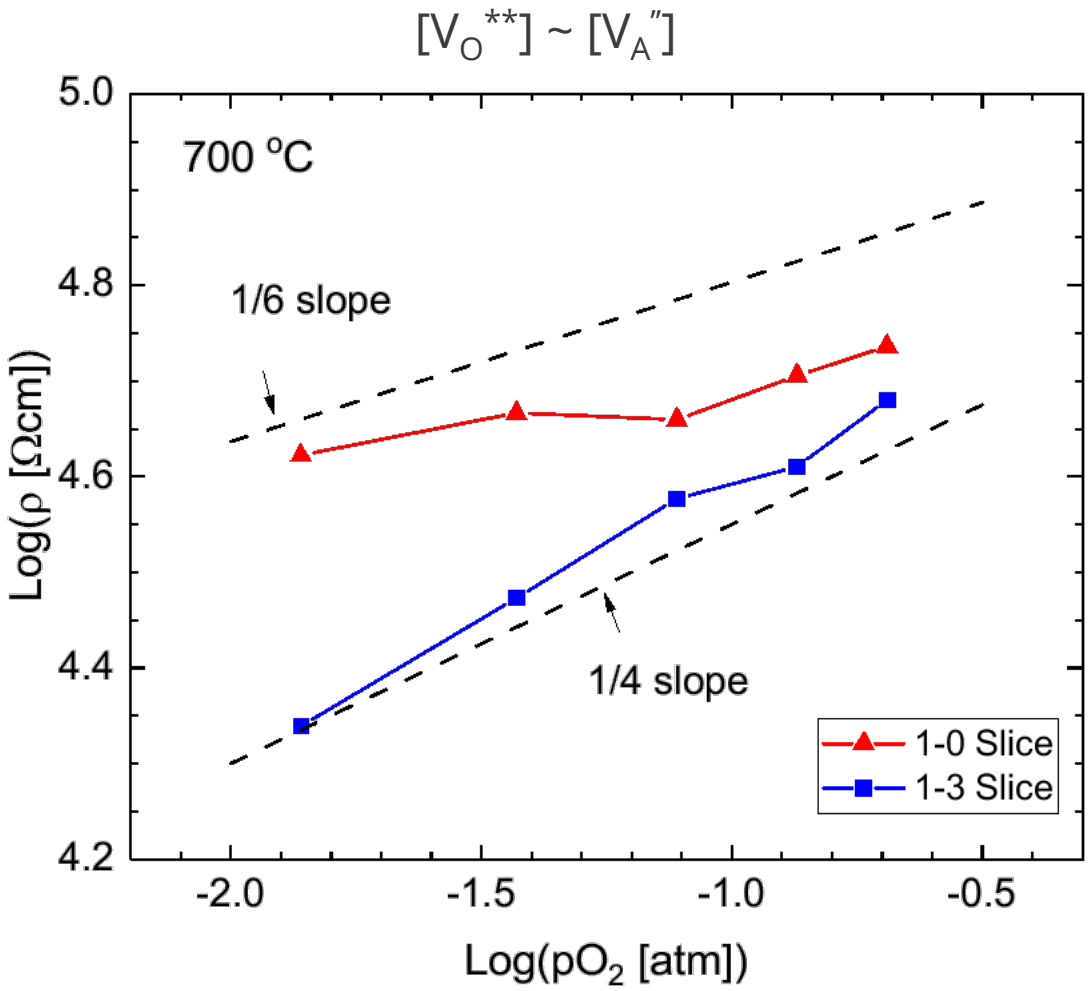




- Thermal annealing dramatically reduces TSDC response, consistent with TSDC probing V_O^{**}
- Larger TSDC in 1-3 slice after anneal

Annealed High Temperature and Oxygen Pressure Dependent Resistivity

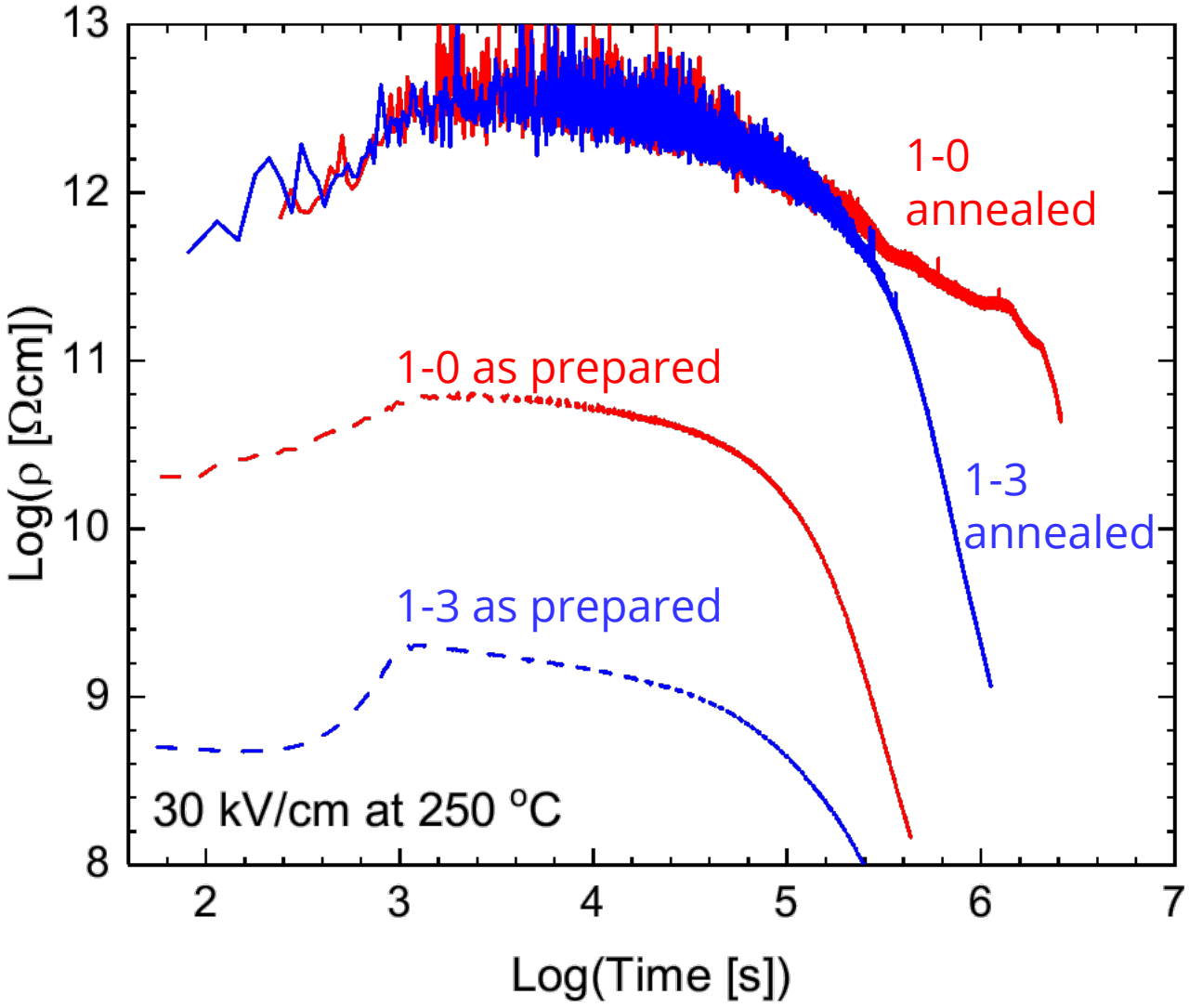




- Temperature dependent resistivity consistent with 20BZT-BT literature[2]
- 1/4 slope oxygen partial pressure dependence consistent with Bi-rich BZT-BT[3]

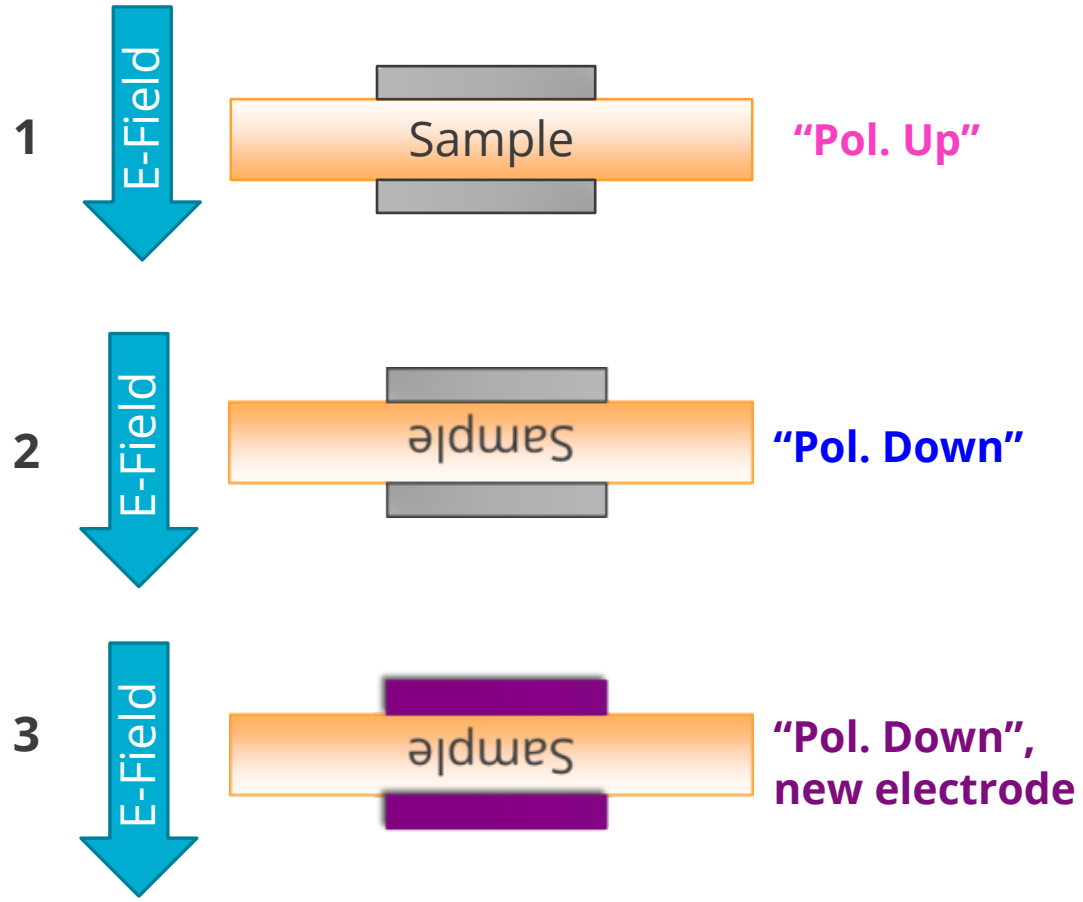
[2]Raengthon et al., Appl. Phys. Lett. 101, 112904 (2012); [3]Kumar et al., J. Am. Ceram. Soc. 101 (2018) 2376

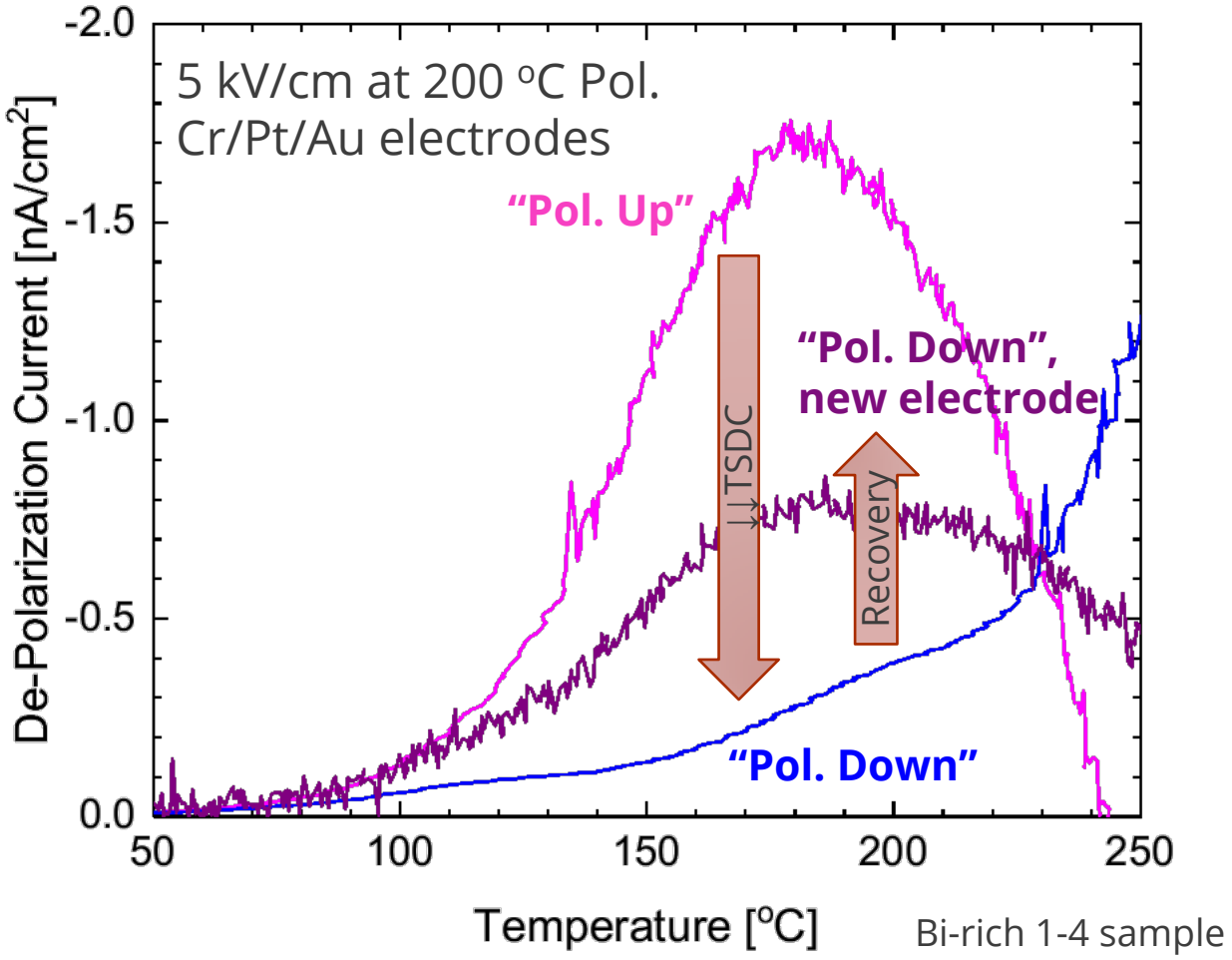
DC Lifetime



→ Sintering in air may cause high [V_o^{**}], reducing DC lifetime

Irreversible Polarity Dependence of TSDC





Bi-rich 1-4 sample

- Irreversible polarity dependence, recoverable with new electrodes → TSDC defects are located at or near electrodes

Summary

- BZT-BT is a potential high temperature, high field ceramic capacitor dielectric for power conversion applications
- TSDC response correlates with existence of oxygen vacancies
- Sintering likely induces large amounts of V_O^{**}
  - Thermal annealing oxidizes samples, reduces TSDC, and extends DC lifetime
- TSDC source appears to be at or near electrodes

Acknowledgements

- DOE Office of Electricity: Transformer Resilience and Advanced Components (TRAC), program manager Andre Pereira
- Clive Randall at Pennsylvania State University