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1. N. Raengthon, T. Sebastian, D. Cumming, I.M. Reaney, and D.P. Cann, "BaTiO<sub>3</sub>-Bi(Zn<sub>1/2</sub>Ti<sub>1/2</sub>)O<sub>3</sub>-BiScO<sub>3</sub> Ceramics for High-Temperature Capacitor Applications," *J. Am. Ceram. Soc.*, 95[11] 3554-3561 (2012). 2. N. Raengthon, V.J. DeRose, G.L. Brennecka, and D.P. Cann, "Defect Mechanisms in High Resistivity BaTiO<sub>3</sub> - Bi(Zn<sub>0.5</sub>Ti<sub>0.5</sub>)O<sub>3</sub> Ceramics," *Appl. Phys. Lett.*, 101, 112904 (2012).

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# CHEMICAL HETEROGENEITY IN ELECTROCERAMICS: THE GOOD, THE BAD, AND THE DIFFICULT TO CHARACTERIZE!

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As characterization techniques continue to advance, the materials community is reminded again and again that our samples are not as perfect as we generally describe them to be. This presentation will focus on  $\text{Bi}(\text{Zn}_{0.5}\text{Ti}_{0.5})\text{O}_3$ - $\text{BaTiO}_3$ -based ceramics in which subtle mesoscale cation gradients have been identified as a key factor in the phenomenal temperature- and field-stable permittivity of these unusual dielectrics as well as their remarkably high resistivity values and associated activation energies.[1,2] Earlier work has shown that the single perovskite phase that results after calcination of mixed oxides and carbonates is formed through a complex series of solid-state reactions (Figure 1),[3] and complementary sintering studies have strongly suggested that development of these complex microstructures with mesoscale heterogeneity is strongly dependent upon cation diffusion kinetics (Figure 2). Here, we report on the effects of reaction pathways during calcination on phase formation and microstructural development during sintering in ceramics of nominally identical  $x\text{Bi}(\text{Zn}_{0.5}\text{Ti}_{0.5})\text{O}_3 - (1-x)\text{BaTiO}_3$  compositions. These results remind us once again that while often treated as such, material micro/meso/nanostructure is not a state function, and that local ion environments can be determined by processing steps, which can in turn profoundly and selectively affect phase formation, ion diffusion, microstructure development, and resultant properties.

This reinforces the need for multiple complementary characterization and measurement techniques for effective description of complex functional materials, and provides a cautionary tale for the budding age of computational materials discovery that real materials—and occasionally enabling performance—often live outside the realm of thermodynamic equilibrium.

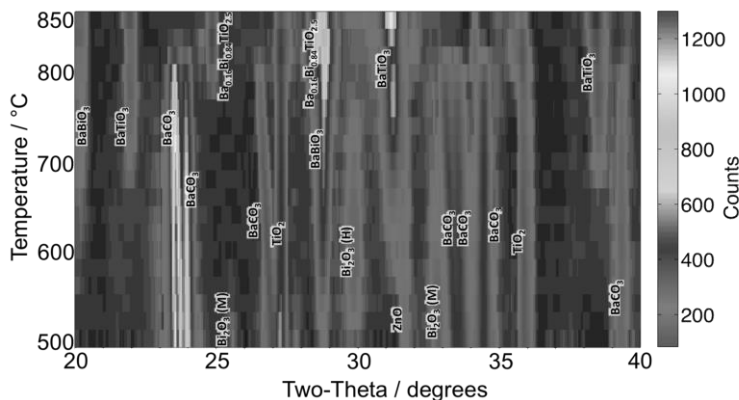


Figure 1 – Temperature-dependent x-ray diffraction during calcination of mixed oxide and carbonate precursors revealed at least four transient intermediate phases before complete conversion to perovskite.[3]

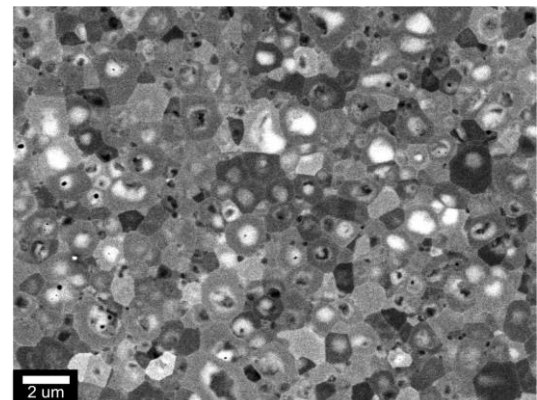


Figure 2 – Highly contrasted backscatter scanning electron microscopy image revealing chemical heterogeneity in  $20\text{Bi}(\text{Zn}_{0.5}\text{Ti}_{0.5})\text{O}_3$ - $80\text{BaTiO}_3$  ceramics.

1. N. Raengthon, T. Sebastian, D. Cumming, I.M. Reaney, and D.P. Cann, "BaTiO<sub>3</sub>-Bi(Zn<sub>1/2</sub>Ti<sub>1/2</sub>)O<sub>3</sub>-BiScO<sub>3</sub> Ceramics for High-Temperature Capacitor Applications," *J. Am. Ceram. Soc.*, **95**[11] 3554-3561 (2012).
2. N. Raengthon, V.J. DeRose, G.L. Brennecka, and D.P. Cann, "Defect Mechanisms in High Resistivity BaTiO<sub>3</sub> - Bi(Zn<sub>0.5</sub>Ti<sub>0.5</sub>)O<sub>3</sub> Ceramics," *Appl. Phys. Lett.*, **101**, 112904 (2012).
3. N. Triamnak, G.L. Brennecka, H.J. Brown-Shaklee, M.A. Rodriguez, and D.P. Cann, "Phase formation of BaTiO<sub>3</sub> - Bi(Zn<sub>1/2</sub>Ti<sub>1/2</sub>)O<sub>3</sub> perovskite ceramics," *J. Ceram. Soc. Jpn.*, **122**[4] 260-266 (2014).