Entropy-Stabilized Oxides

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The hunt for, and discovery of, new phases of matter is among the most exciting activities Materials Research. The excitement originates from the possibility of active property engineering and advanced technology opportunities that serve societal needs. As a community, we have been looking for a long time, and given the finite number of practical elements and their stable combinations, it is not clear how many are left to find especially if we limit ourselves to conventional tactics. In response, researchers are exploring new approaches that include artificial layering, high throughput computation, severely confined dimensions, and strain-stabilization.

This presentation will explore the concept of entropy engineering, where many cations are distributed at random among a single sublattice creating a high degree of configurational disorder. At finite temperatures, these systems can minimize free energy by remaining single phase. While such processes are known in nature, *i.e*., the energetics that regulate site occupancy in spinels, they were adopted recently by the metallurgy community to create a new class of engineering alloys with exciting properties.

At NCSU, this concept was extended to complex oxides to demonstrate without ambiguity that new phases of matter can be stabilized by configurational disorder. These crystalline materials are referred to as Entropy-Stabilized Oxides (ESOs) and feature local coordination values that are uncommon in natural and synthetic materials.

Using a series of solid-state chemistry experiments, a simple thermodynamic model, and a five-component oxide formulation, we demonstrate beyond reasonable doubt that entropy can predominate the thermodynamic landscape, and drive a reversible solid-state transformation between multiphase and single phase states, where in the latter, cation distributions are random and homogeneous. The findings validate the hypothesis that deliberate configurational disorder provides an orthogonal strategy to imagine new phases of crystalline matter and exploit new opportunities for property engineering.

Finally, we will demonstrate how the palette of entropy-stabilized oxides can be expanded when the formulations are prepared using physical vapor deposition, *i.e*., the range of compositions that become entropy stabilized increase substantially upon thin film preparation as compared to bulk solid-state synthesis. At this stage we speculate that kinetic energy of the incoming species provides an effective temperature that increases the entropic contribution to free energy. This methods leads to even more unusual cation accommodation and immediate possibilities for lattice constant engineering beyond the limits of conventional solid solution.

ABOUT THE SPEAKER: Jon-Paul Maria is a Professor of Materials Science and Engineering at North Carolina State University, and an NCSU University Faculty Scholar. Jon-Paul Graduated from The Pennsylvania State University with B.S., M.S., and Ph.D. degrees in Ceramic Science in 1994, 1996, and 1998, respectively. Jon-Paul and his research group specialize in novel materials development, synthesis, and in tegration science, with a general focus on electronic oxides. Jon-Paul currently directs or co-directs research programs on oxide-nitride heterostructures, Mid-IR plasmonic materials, ferroelectric thin films, nano-energetics, and entropy-stabilized oxides, and extreme high temperature refractories. Research activities of note from the Maria Group include the exploration of entropy-stabilized complex oxides, extreme-mobility transparent conductors, novel approaches for thin film packaging, understanding scaling effects in ferroelectric thin films, and epitaxial integration of functional oxides with wide band gap semiconductors.