High-Entropy Oxides: Multifunctionality Enabled through Chemical Disorder

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Abstract: Entropy engineering offers access to a vast, unexplored materials discovery space by utilizing configurational entropy as a thermodynamic driver towards new multicomponent crystalline solids. Our interdisciplinary team within the Materials Research Science and Engineering Center (MRSEC) at Penn State explores high-entropy oxide (HEO) materials in which the cation sublattices are occupied by many elements at random. The ability to stabilize elements in unusual states within the high-entropy matrix offers an attractive opportunity for functional property engineering in complex oxides, specifically ionic and transparent conductors, relaxor ferroelectrics, and strongly correlated materials. We have recently discovered that entropy-significant states can be controlled within the prototypical HEO, (MgCoNiCuZn)O, through tuning of kinetic variables during thin film synthesis. Advanced microscopy techniques assisted by unsupervised machine learning were used to identify two nanostructures within the thin film, a copper "tweed" and spinel "nanocuboids". Density functional theory calculations and phase-field modeling were able to validate these results as well as simulate the formation of the copper "tweed" phase. We hypothesize that such structures "withdraw" from their entropy bank to acquire these nanostructures, opening a new pathway of HEO materials discovery. A variety of other oxide crystal structures are being investigated for property specific engineering, such as rare-earth fluorites for fast-ion conduction, perovskites for transparent conduction and ferroelectricity, and single-crystal magnetic systems for spintronic applications. As improvement of such properties commonly leads to decreased thermodynamic stability, transition-metal and rare-earth oxide materials offer excellent opportunities for employing an entropy-driven approach. Our team's efforts hope to push the materials discovery frontier beyond the classical realm of enthalpic stability by utilizing entropy for the next generation of complex-oxide materials.