Discovering High-Entropy Oxides with a Machine-Learning Interatomic Potential

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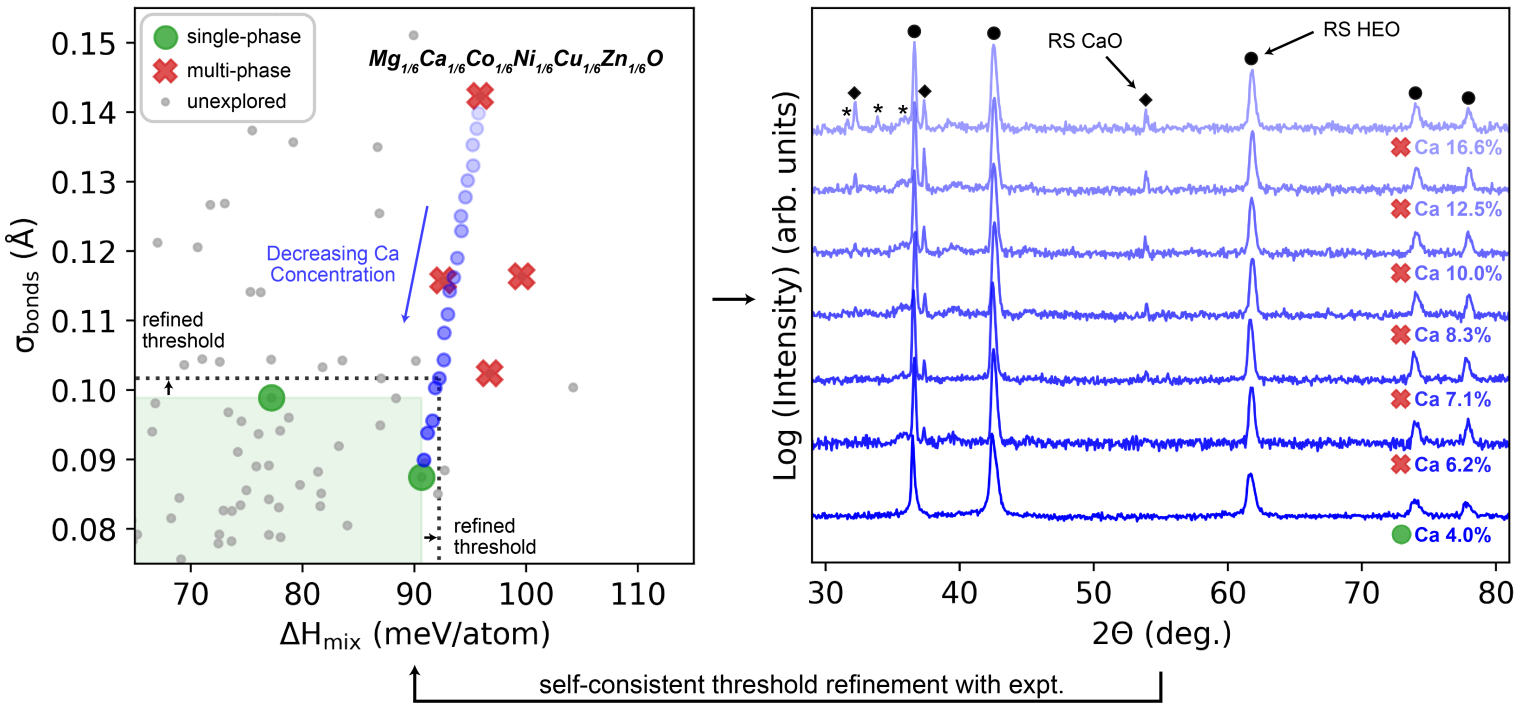
High-entropy materials shift the traditional materials discovery paradigm to one that leverages disorder, enabling access to unique chemistries unreachable through enthalpy alone. A MRSEC team has developed a high-throughput framework for discovering and understanding the single-phase formation of high-entropy oxides (HEOs) by integrating computation and experiment in a self-consistent feedback loop. To more rapidly explore rock salt composition space, the team utilizes CHGNet machine-learning interatomic potentials with impressive accuracy even in disordered systems.

Two computational descriptors resolve the single-phase stability for all eight equimolar rock salt HEO compositions explored to date and lead to the discovery of a novel non-equimolar rock salt HEO containing Ca. This collaborative workflow workflow is currently being applied to explore more complex crystal structures that may possess emerging property opportunities.



Left: Computational phase map of rock salt HEO composition space quantified using the mixing enthalpy (ΔH_{mix}) and standard deviation of bond lengths (σ_{bonds}).

Right: X-ray diffraction experimental with decreasing Ca% for $Mg_{1/6}Ca_{1/6}Co_{1/6}Ni_{1/6}Cu_{1/6}Zn_{1/6}O$. Black line shows a refined single-phase stability threshold.