Thermodynamics-Inspired High Entropy Oxide Synthesis

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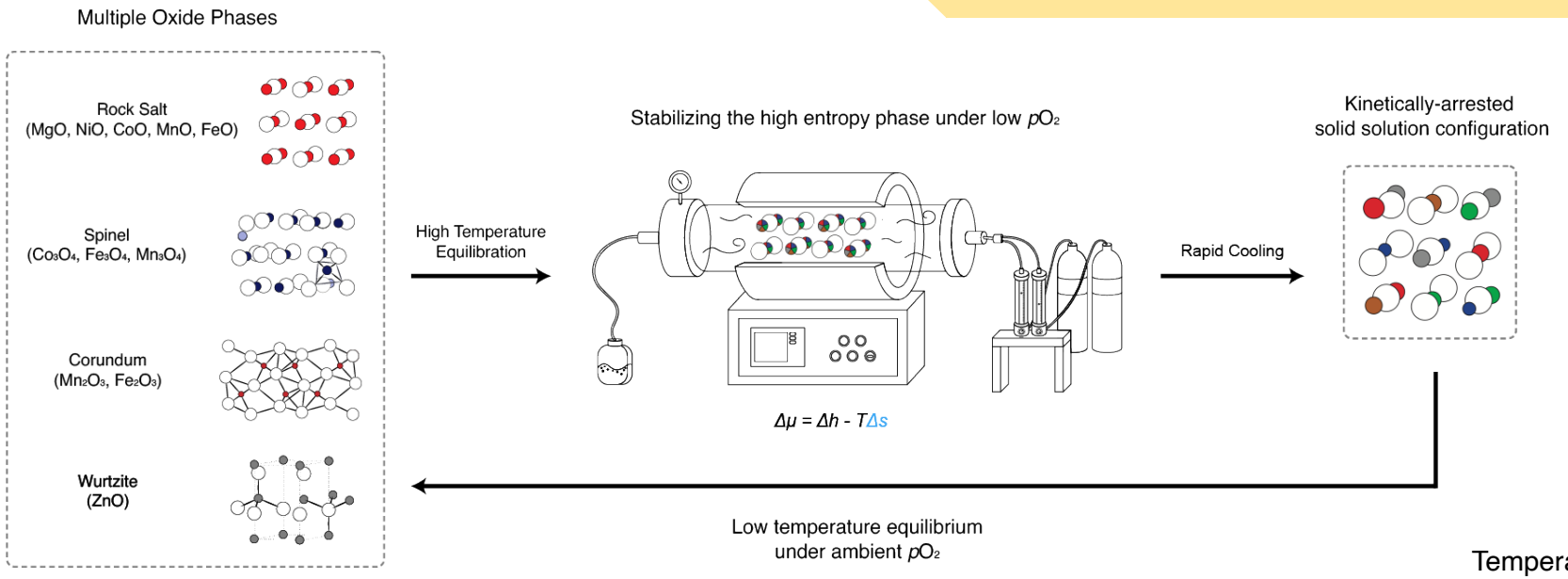

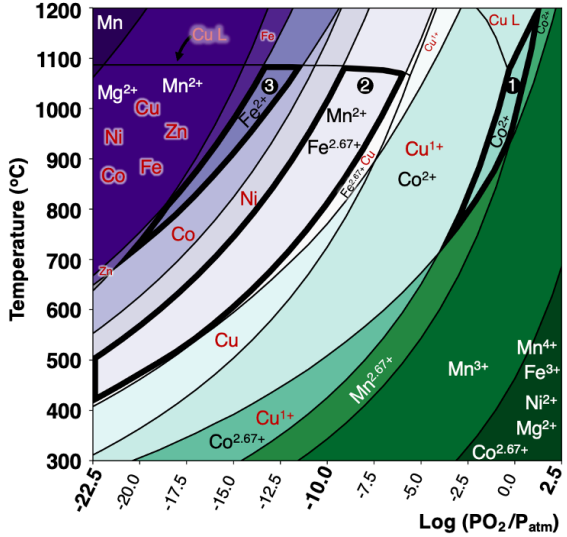

Temperature and oxygen partial pressure phase diagram illustrating each cation's preferred oxidation state in its stable binary oxide.

A MRSEC team has constructed phase diagrams to predict entropy-stabilized materials, thus guiding the control of oxygen levels during synthesis to incorporate Mn^{2+} and Fe^{2+} into rock salt high-entropy oxides — a structure where they are typically unstable in air. This work establishes design rules for metastable oxides that host multivalent cations with novel functionality. Supported by the IRG's "Ceramic Factory" undergrads, the team has synthesized seven equimolar single-phase rock salt compositions that accommodate multivalent Mn, Fe, or both, as confirmed by X-ray diffraction and fluorescence. X-ray absorption fine structure spectra reveal predominantly divalent cations. Although the initial focus is on rock salt HEOs, these methods are chemically and structurally agnostic, providing a broadly adaptable framework for navigating HEOs thermodynamics and enabling a broader compositional range with contemporary property interest.