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Machine-Learning-driven discovery of High-Entropy Metal Phosphates and the mechanistic Insights into their crystallisation pathways

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High-entropy metal phosphates (HEMPs) are a class of solid solutions that incorporate at least five metal cations into a crystal lattice and are thereby stabilised by entropy of mixing [1]. HEMPs are analogous to naturally occurring minerals such as struvite, dittmarite, and vivianite, which frequently form in engineered environments during wastewater treatment or soil remediation. Moreover, due to their structural versatility, HEMPs are investigated for electrochemical applications. Controlling the synthesis and phase stability of these solid solutions requires a fundamental understanding of their formation pathways.

Our in situ scattering and electron microscopy investigations reveal that the formation of metal phosphates is non-classical [2,3]. The early stages of these processes involve liquid-liquid phase separation and the formation of transient, colloidal amorphous nanophases, preceding crystallisation. In mixed-metal systems, such as Ni-Co phosphates, the nucleation and crystallisation pathways become significantly convoluted. Rather than following a predictable interpolation of pure endmembers, multicationic mixtures exhibit prolonged metastable phase coexistence and elemental fractionation. The complexity of these nanoscale amorphous-to-crystalline transformations makes navigating the vast compositional space to discover novel HEMPs a challenge. To address this, we developed a workflow integrating automated wet-chemical synthesis with sequential learning predictions. By deploying a Random Forest regression model, we iteratively evaluated targeted experimental outcomes to efficiently explore untested formulations. This data-driven approach successfully predicted the formation of novel solid solutions and guided the synthesis of HEMPs incorporating up to seven distinct cations.

Yet, the success of statistical methods in navigating this compositional space highlights a deeper scientific challenge. While data-driven frameworks excel at accelerating macroscopic discovery, the underlying structural and chemical intricacies of nucleation and crystallisation remain difficult to predict. Achieving a truly predictive understanding of complex geochemical reactions and mineral-fluid systems requires pairing ML algorithms with rigorous fundamental research into non-classical mechanisms.

[1] Karafiludis, Standl, Ryll, Schwab, Prinz, Wolf, Kruschwitz, Emmerling, Völker & Stawski (2025), *Crystal Growth & Design* 25, 7989-8001.

[2] Karafiludis, Kochovski, Scoppola, Retzmann, Hodoroaba, ten Elshof, Emmerling & Stawski (2023), *Chemistry of Materials* 35, 10645-10657.

[3] Karafiludis, Scoppola, Wolf, Kochovski, Matzdorff, Van Driessche, Hövelmann, Emmerling & Stawski (2023), *The*