Disordered High-Entropy Transition-metal Hydroxides: Synthesis, Local Structure and OER Activity

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Recent studies have shown that introducing atomic-scale disorder in oxide and hydroxide materials can enhance their properties as oxygen evolution reaction (OER) electrocatalysts. However, despite this growing evidence that disordered, earth-abundant materials are promising alternatives to benchmark noble-metal catalysts, their local atomic structure remains poorly understood [1-2]. In particular, the absence of long-range order limits the structural information accessible by conventional diffraction techniques. Establishing controllable synthesis strategies also remains a key challenge for rational materials design. In this project, we investigate how composition and synthesis conditions direct the local atomic motifs in disordered high-entropy (HE) transition-metal (TM) hydroxides using X-ray total scattering with Pair Distribution Function (PDF) analysis. Using hydrothermal synthesis [3], disordered HE TM hydroxides containing Mn, Fe, Co, Ni and Cu were prepared, together with mono-, bi- and tetrametallic compositions to identify how the incorporated 3d metals influence the structure of the final disordered product (Figure 1). Preliminary PDF analysis shows that the tetrametallic and HE samples exhibit features similar to those of layered double hydroxides (LDH) [4], while the bimetallic samples display local structural correlations resembling spinel-type motifs (Figure 1b and 1c). Initial evaluation of the OER electrocatalytic activity shows promising performance. Ongoing work focuses on detailed structural modelling and correlating the local atomic structure with catalytic activity in these disordered HE TM hydroxides.

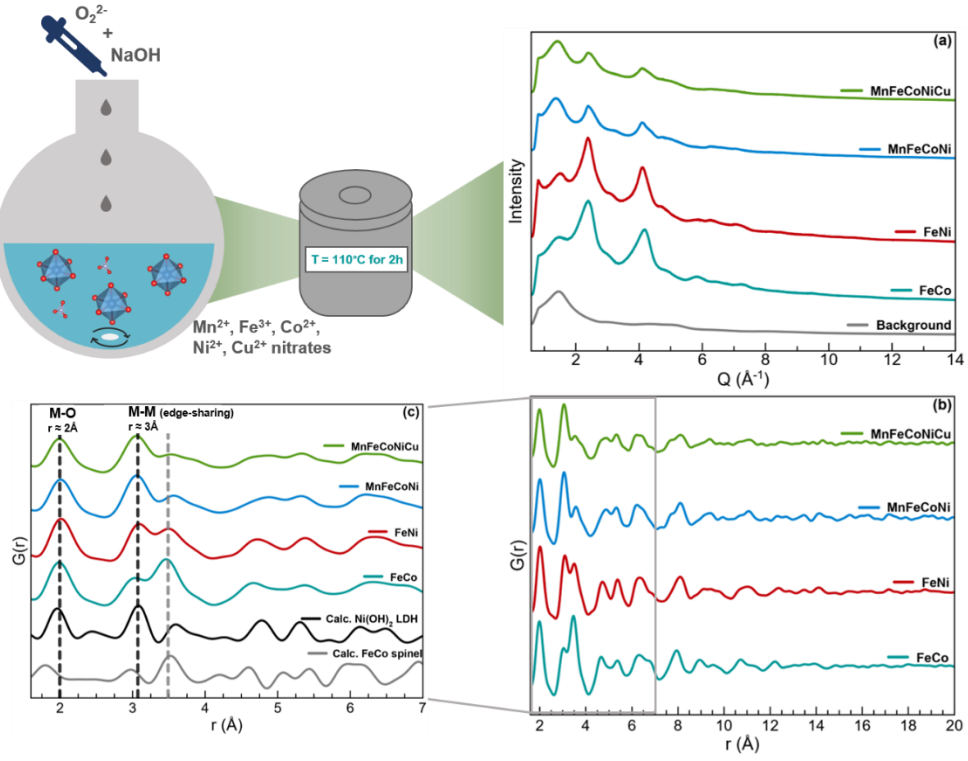


Figure 1. *Left: Scheme of the hydrothermal synthesis. Right: (a) X-ray total scattering data measured on in-house diffractometer (λ=0.56Å). (b) Corresponding Pair Distribution Functions (Q_{max}=17Å). (c) Fingerprinting experimental PDFs with calculated Ni-LDH (R-3mH) and FeCo spinel (Fd-3m) PDFs.*

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