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Title: Discovery of High-Entropy Oxide Electrocatalysts: From Thin- FilmMaterial Libraries to Particles

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Abstract: Discovery of new high-entropy electrocatalysts requires testing of hundreds to thousands of possible compositions, which can be addressed most efficiently by high-throughput experimentation on thin-film material libraries. Since the conditions for high-throughput measurements ("screening") differ from more standardized methods, it is frequently a concern whether the findings from screening can be transferred to the commonly used particulate catalysts. We demonstrate the successful transfer of results from thin-film material libraries to particles of Cantor alloy oxide (Co-Cr-Fe-Mn-Ni)₃O₄. The chemical compositions of the libraries, all singlephase spinels, cover a wide compositional range of (Cr_{8.1-28.0}Mn_{11.6-28.4}-Fe_{10.6-39.0}Co_{11.4-36.7}Ni_{13.5-31.4})_{37.7} +/- 0.6O_{62.3} +/- 0.6, with composition-dependent lattice constant values ranging from 0.826 to 0.851 nm. Electrochemical screening of the libraries for the oxygen evolution reaction (OER) identifies (Cr_{24.6} +/- 1.4- Mn_{15.7} +/- 2.0Fe_{16.9} +/- 1.8Co_{26.1} +/- 1.9Ni_{16.6} +/- 1.7)_{37.8} +/- 0.8O_{62.2} +/- 1.2 as the most active composition, exhibiting an overpotential of 0.36 V at a current density of 1 mA cm⁻². This "hit" in the library was subsequently synthesized in the form of particles with the same composition and crystal structure using an aerosol-based synthesis strategy. The similar OER activity of the most active thin-film composition and the derived catalyst particles validates the proposed approach of accelerated discovery of novel catalysts by screening of thin-film libraries.

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