

Searching novel complex solid solution electrocatalysts in unconventional element combinations

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The composition spreads for each element, which result from the 45° tilted orientation of each cathode relative to the substrate surface, are shown in Figure S1. Since Ni and Ti are sputtered from a single alloy target, their concentration gradients are identical.

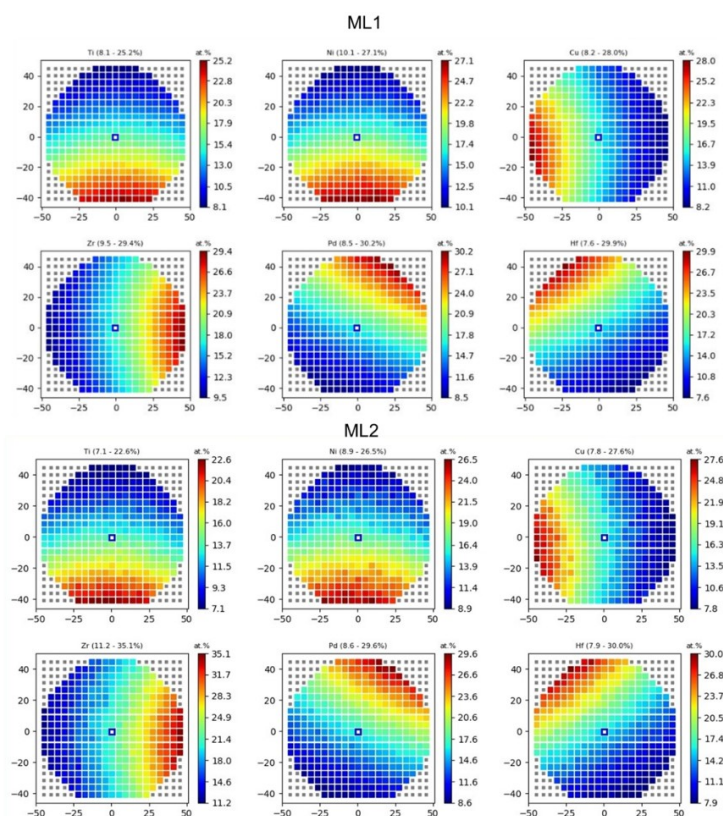


Figure S1. EDS composition maps for each element in the combinatorial materials library. Note that TiNi46.9 come from a single alloy target, resulting in an identical distribution pattern. The indicated center composition for ML1 is Ti 16 at.%, Ni 19 at.%, Cu 16 at.%, Zr 18 at.%, Pd 16 at.%, Hf 15 at.%. The center composition for ML2 is Ti 14 at.%, Ni 17 at.%, Cu 16 at.%, Zr 21 at.%, Pd 17 at.%, Hf 15 at.%.

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XRD (Figure S2) of the as-deposited state revealed no discernable diffraction peaks, with only a broad hump located around approximately $40^\circ 2\theta$, indicative of amorphous material.

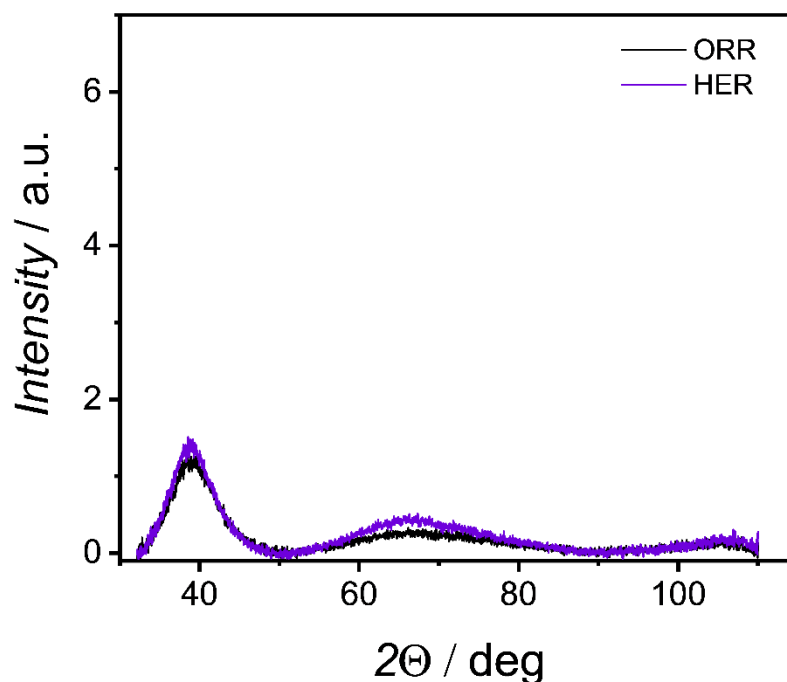


Figure S2. Representative XRD patterns indicating amorphous structure in the as-deposited state. Position 168 corresponds to the center of the most ORR active region, while Position 248 is in the center of the most HER active region, indicated in Figure 1 of the main text.

Confirmation of reliability of electrochemical measurements:

In order to exclude any systematic error introduced by the use of automated measuring setup, the ML was measured a second time after being physically rotated by 90° . The obtained results, changes of activity, changed along with turning the sample are confirming that the observed changes in activity are caused by the change in composition of the ML.

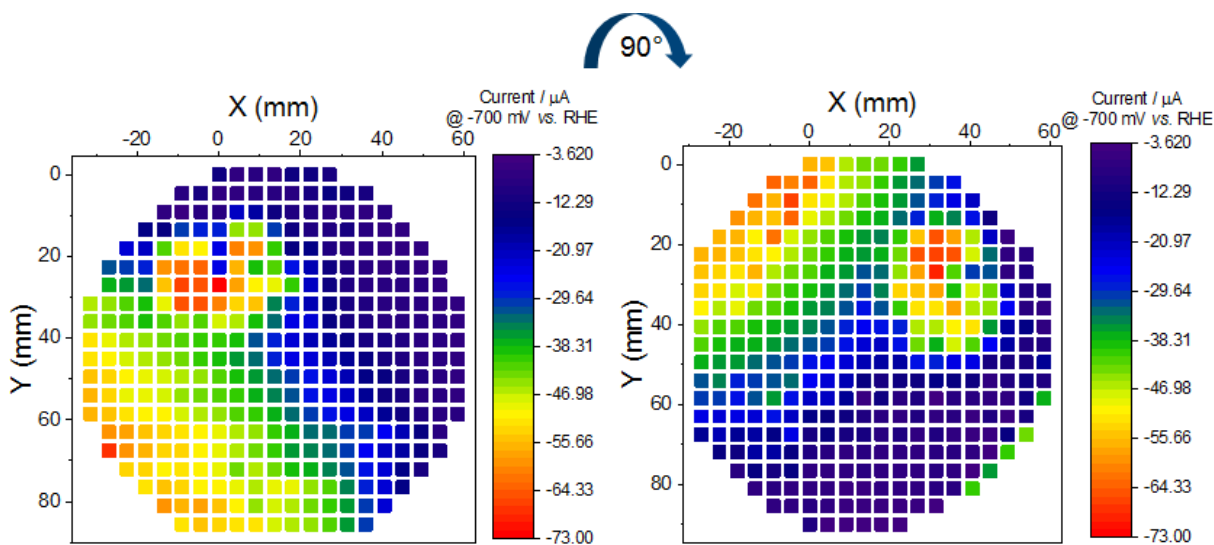


Figure S3. Electrochemical activity maps obtained by plotting the current at -700 mV vs. RHE measured on ML initially, and then subsequently after physical rotation of the sample by 90° .

Hydrogen evolution reaction activity maps at different potentials:

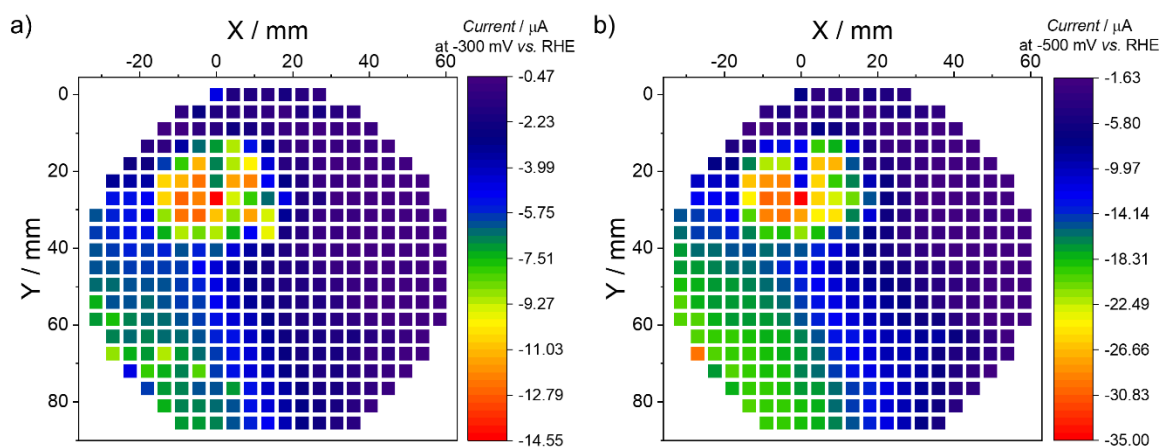


Figure S4. HER activity maps of ML2 at a) -300 mV vs. RHE and b) -500 mV vs. E.