

**Supplementary Information**

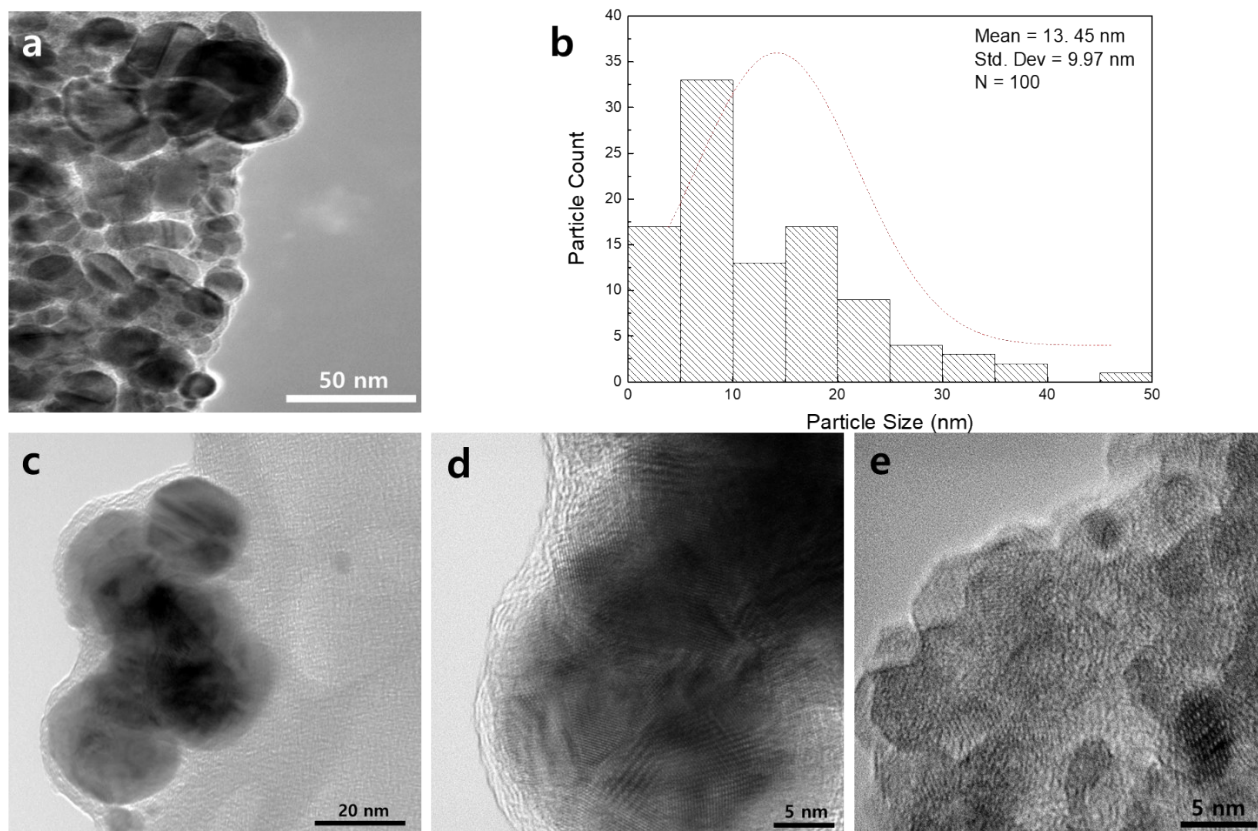
**Universal pH Electrocatalytic Hydrogen Evolution by Au-based High Entropy Alloys**

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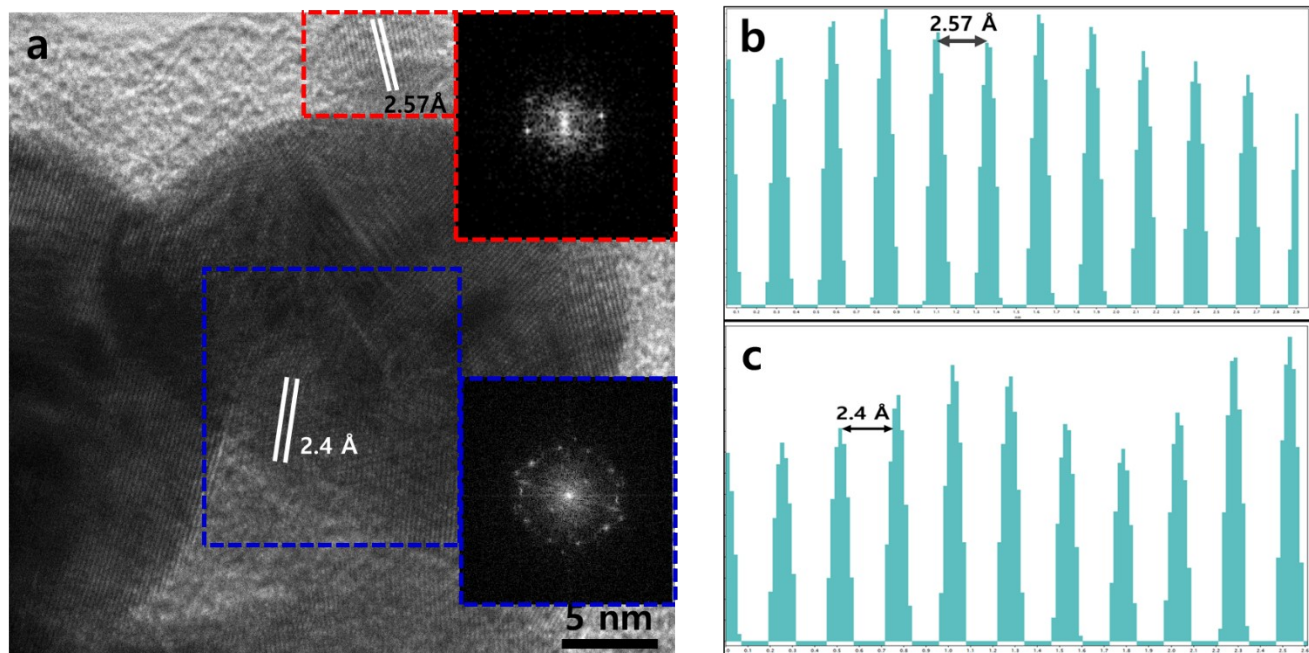
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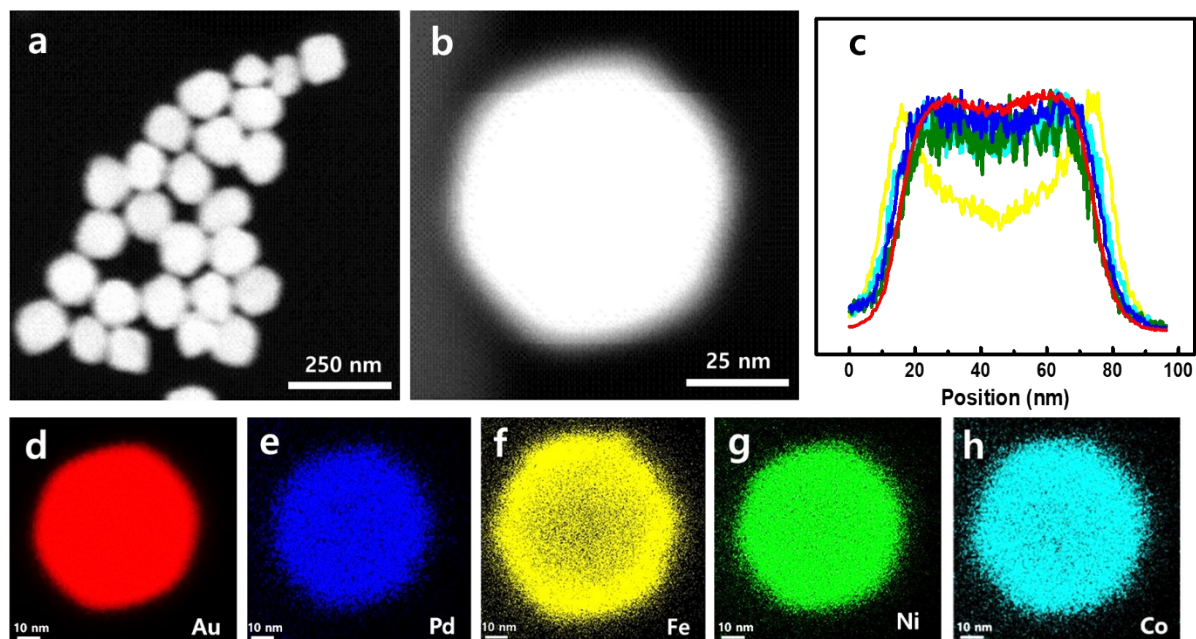
\* E-mail: [Michael\\_Ross@uml.edu](mailto:Michael_Ross@uml.edu) (Ross, Michael B).



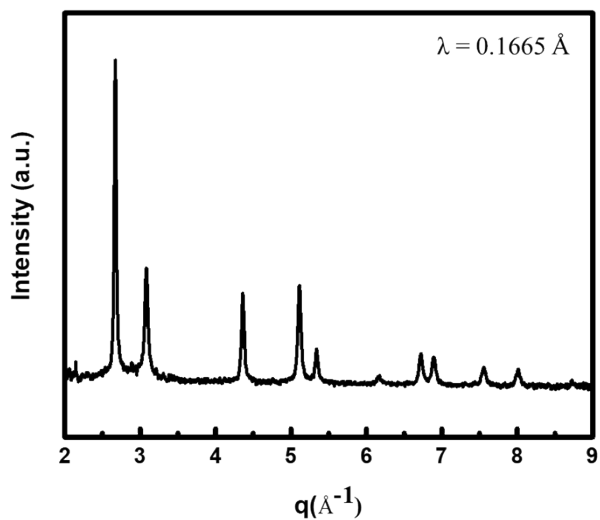
**Fig. S1** High resolution-TEM (HR-TEM) images (a) representative HR-TEM image of AuPdFeNiCo HEA NPs, (b) particle size distribution histogram of AuPdFeNiCo HEA NPs, HR-TEM characterization of AuPdFeNiCo HEAs with different magnification (c), (d), (e).



**Fig. S2** HR-TEM characterization of AuPdFeNiCo HEA NPs (a) FFT images of AuPdFeNiCo HEAs corresponding high resolution-TEM (HR-TEM) image of the area enclosed by the insert box. (red:  $\text{Fe}_3\text{O}_4$ , blue: AuPdFeNiCo HEAs) (b) The intensity line profile along the lattice in the red box. (c) The intensity line profile along the lattice in the blue box.



**Fig. S3** STEM-EDS characterization of AuPdFeNiCo HEA NPs (a) Representative scanning transmission electron microscopy (STEM) image and (b) zoomed view of STEM image. (c) crossline profile for all five elements of the corresponding STEM image and energy dispersive spectroscopy (EDS) elemental mapping of (d) Au, (e) Pd, (f) Fe, (g) Ni, and (h) Co.



**Fig. S4** Synchrotron wide-angle X-ray scattering (WAXS) of AuPdFeNiCo HEA NPs.

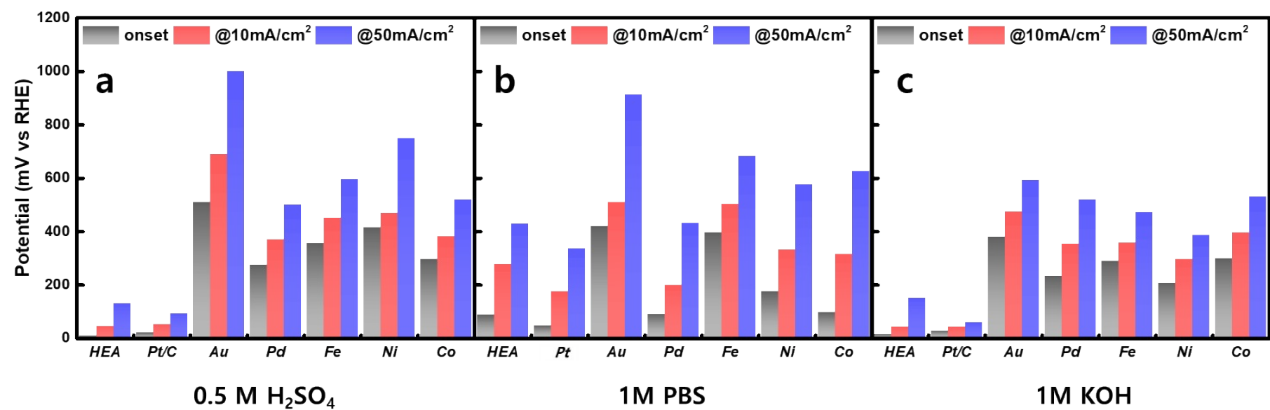


Fig. S5 Summary of HER activity of electrocatalysts in different electrolytes.

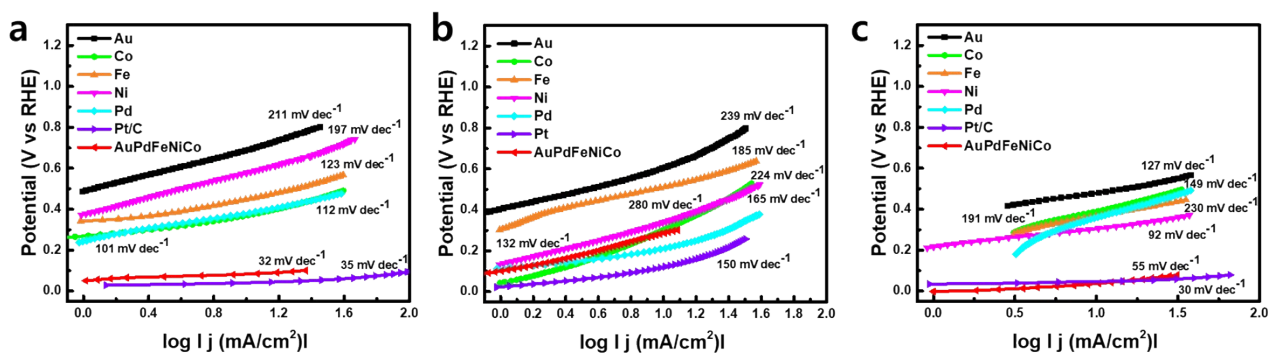
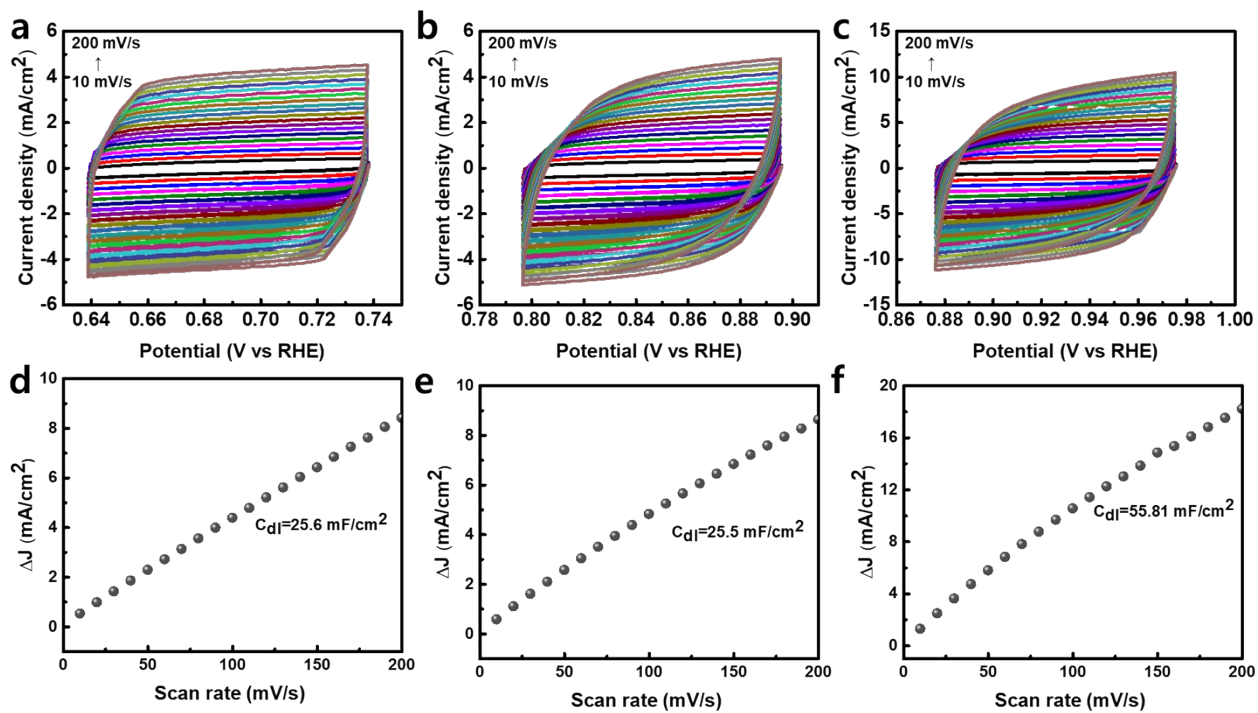
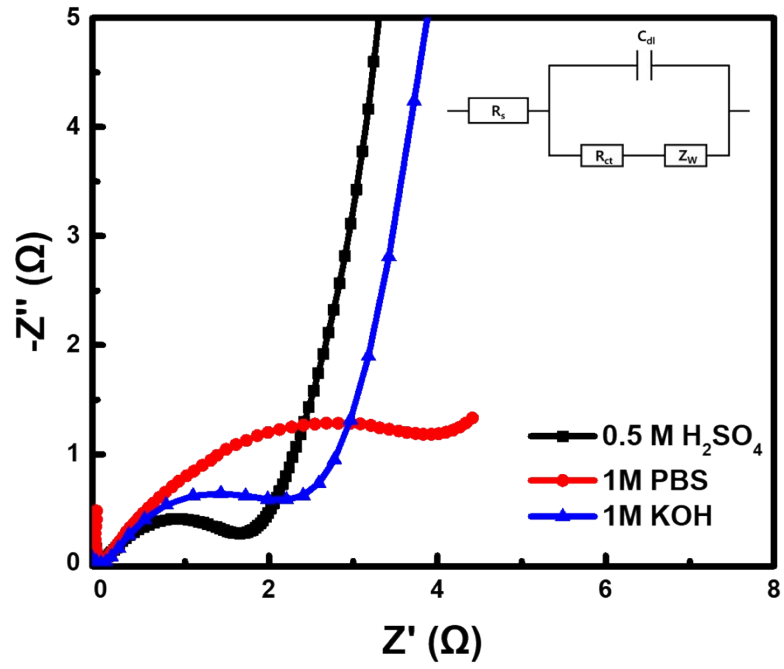


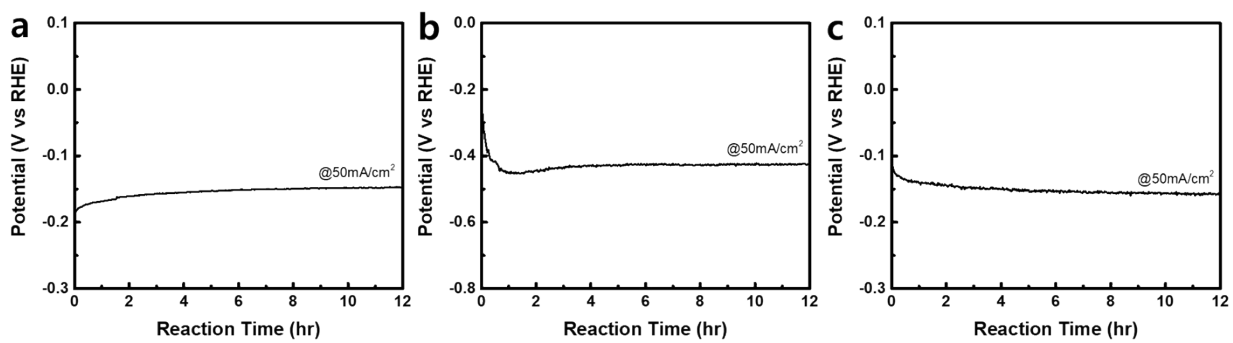
Fig. S6 Corresponding Tafel slope of Au, Pd, Fe, Ni, Co, AuPdFeNiCo, and Pt/C (a) 0.5M H<sub>2</sub>SO<sub>4</sub>, (b) 1M PBS, (c) 1M KOH



**Fig. S7** CV curves at different scan rates from 10 to 200 mV s<sup>-1</sup> in the corresponding electrolytes for AuPdFeNiCo, and fitted C<sub>dl</sub> based on CV, (a), (d) 0.5M H<sub>2</sub>SO<sub>4</sub>, (b), (e) 1M PBS, and (c), (f) 1M KOH.

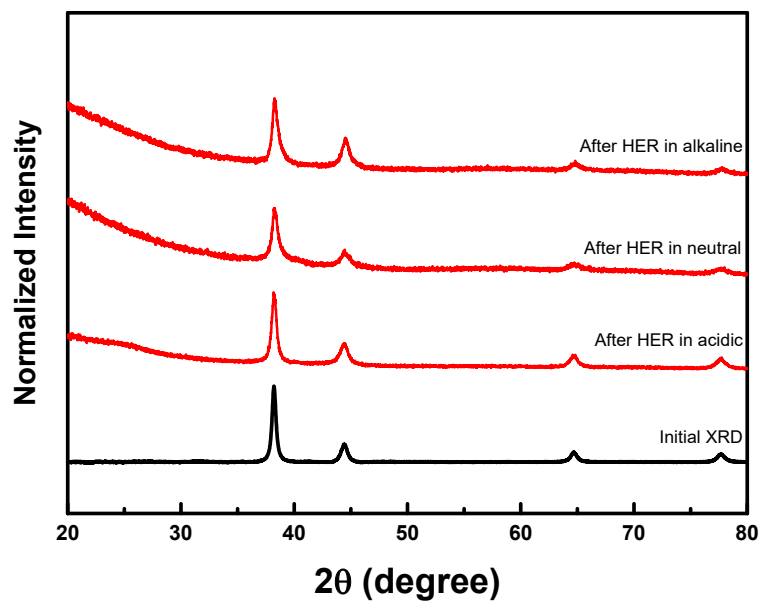


**Fig. S8** The equivalent circuit model for the EIS plots of AuPdFeNiCo HEAs.



**Fig. S9** HER-stability test of AuPdFeNiCo HEAs with chronopotentiometry curve, (a) 0.5M H<sub>2</sub>SO<sub>4</sub>, (b) 1M PBS, and (c) 1M KOH.





**Fig. S10** X-ray diffraction (XRD) characterization of AuPdFeNiCo HEA NPs before and after 12 hr of continuous HER operation in different pH-electrolytes.

**Table. S1** Composition of AuPdFeNiCo HEAs determined by inductively coupled plasma – optical emission spectrometry (ICP-OES).

	Au	Pd	Fe	Ni	Co
Mass loading (wt. %)	1.524	2.448	1.0712	0.684	1.1784
Atomic percentage (at.%)	22.069	35.449	15.512	9.906	17.064