

## Supporting Information

### **Multi-heterostructure with multi-interface for efficient hydrogen evolution reaction towards industrial alkaline seawater**

Kai Zhang,<sup>a</sup> Qingmei Su,<sup>\*b</sup> Yvjie Lv,<sup>a</sup> Weihao Shi,<sup>a</sup> Pinpin Wang,<sup>c</sup> Zhiyong Wang,<sup>b,d</sup> Wenqi Zhao,<sup>b</sup> Miao Zhang,<sup>b</sup> Shukai Ding,<sup>b</sup> Shufang Ma,<sup>b</sup> Gaohui Du,<sup>\*b</sup> and Bingshe Xu<sup>b,c</sup>

<sup>a</sup> School of Materials Science & Engineering, Shaanxi University of Science and Technology, Xi'an, 710021, P. R. China.

<sup>b</sup> Materials Institute of Atomic and Molecular Science, School of Physics & Information Science, Shaanxi University of Science and Technology, Xi'an, 710021, P. R. China.

<sup>c</sup> Key Laboratory of Interface Science and Engineering in Advanced Materials, Taiyuan University of Technology, Taiyuan, 030024, P. R. China

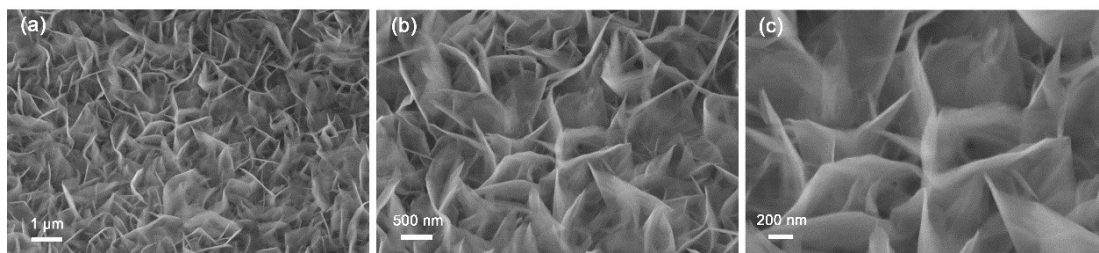
<sup>d</sup> Beijing University of Technology, Chaoyang District, Beijing, 100124, P. R. China

<sup>e</sup> College of Food Science and Pharmaceutical Engineering, Zaozhuang University, Zaozhuang, 277160, P. R. China

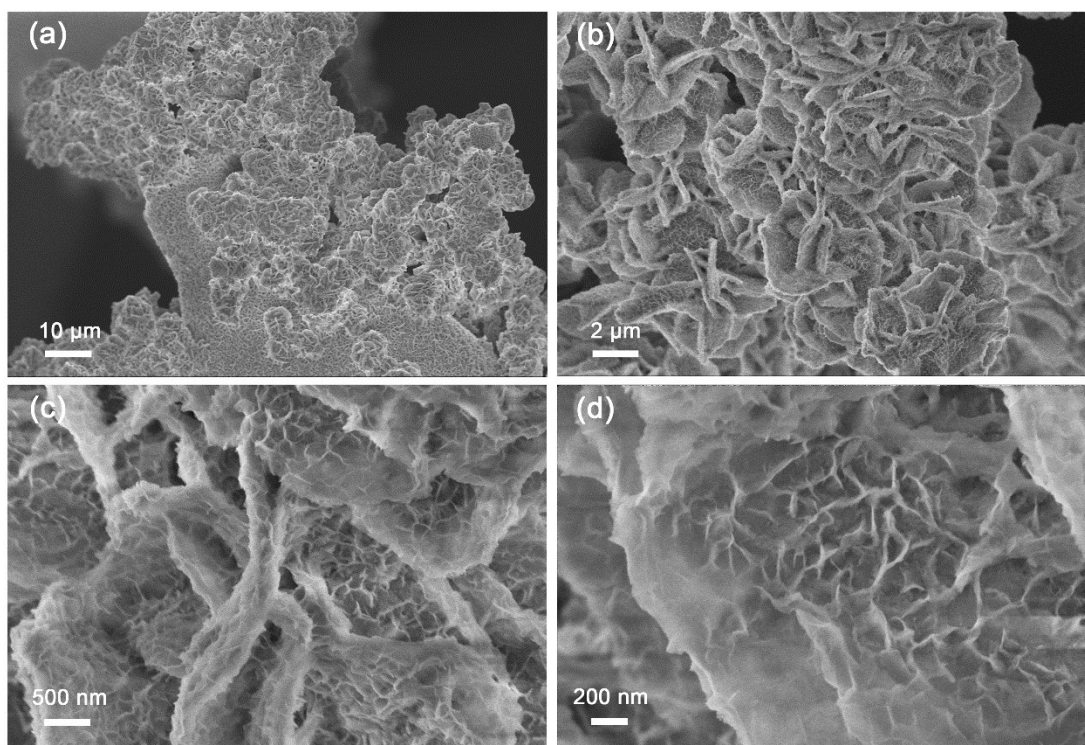
#### **Corresponding authors:**

*E-mail addresses:* suqingmei@sust.edu.cn (Q. Su), dugaohui@sust.edu.cn (G. Du)

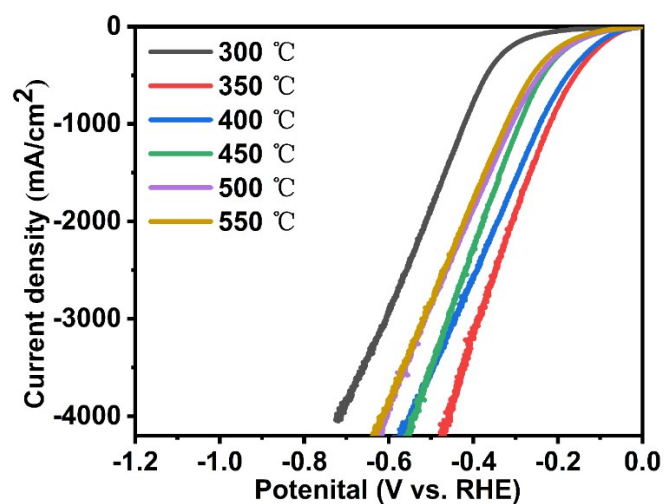
## Supporting Figures



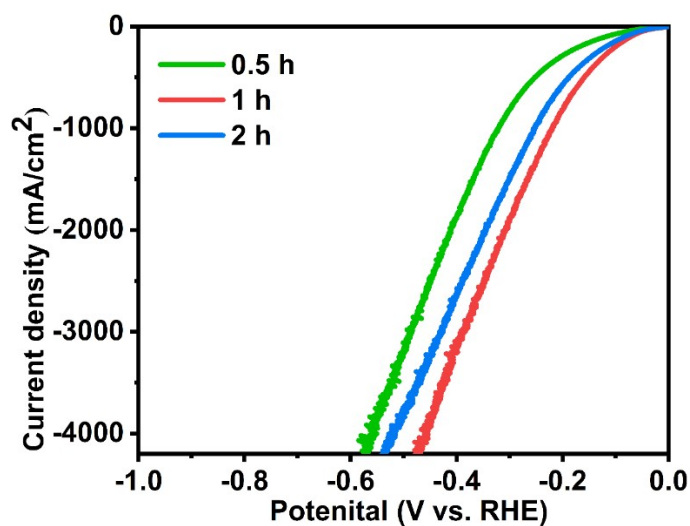
**Figure S1.** The SEM images of  $\text{NiCo}_2\text{O}_4$  nanosheet arrays on the Ni foam.



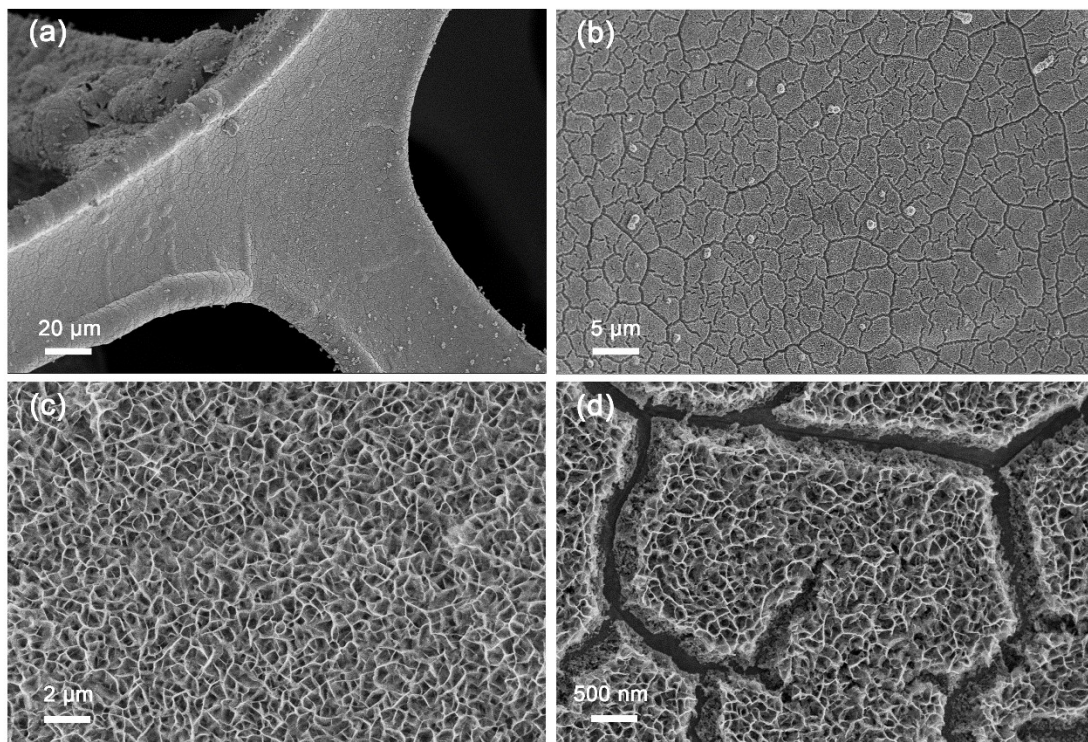
**Figure S2.** The SEM images of the  $\text{NiCo}_2\text{O}_4@ \text{NiMoO}_4$  on the Ni foam.



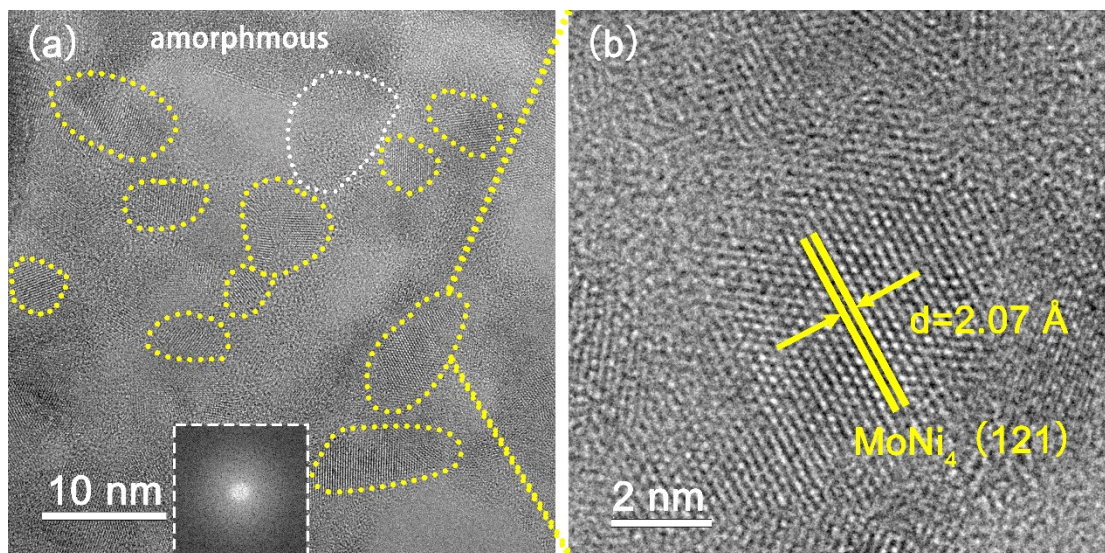
**Figure S3.** The LSV polarization curves with 90% iR compensation of the H<sub>2</sub>-NiCo<sub>2</sub>O<sub>4</sub>@NiMoO<sub>4</sub> nanosheets calcined at different reduction temperatures under H<sub>2</sub>/Ar atmosphere in 1.0 M KOH solution.



**Figure S4.** The LSV polarization curves with 90% iR compensation of the H<sub>2</sub>-NiCo<sub>2</sub>O<sub>4</sub>@NiMoO<sub>4</sub> nanosheets calcined at 350 °C for different reduction times under H<sub>2</sub>/Ar atmosphere in 1.0 M KOH solution.

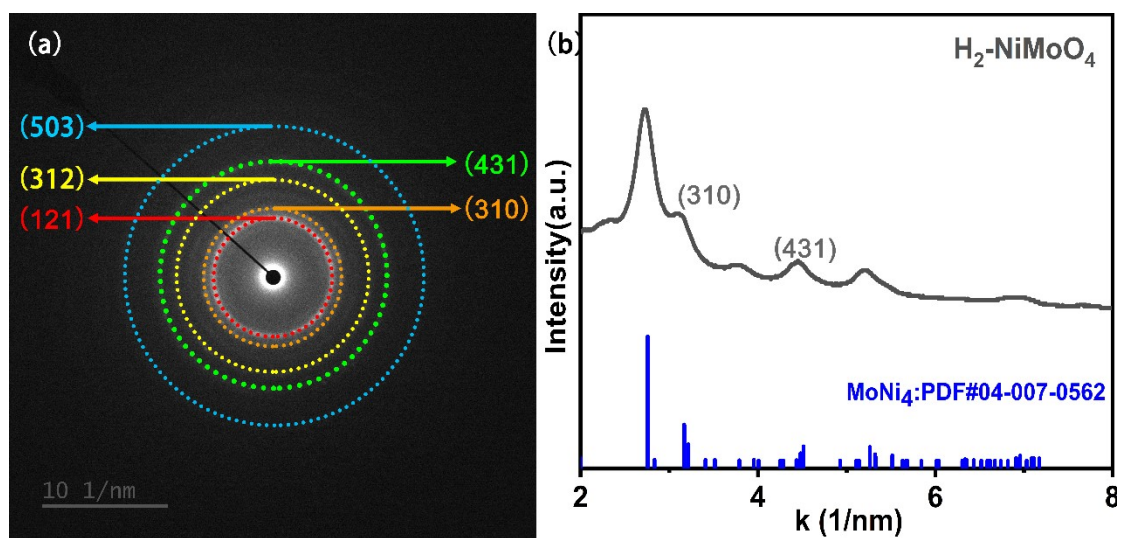


**Figure S5.** The SEM images of  $\text{H}_2\text{-NiMoO}_4$  nanosheet arrays on the Ni foam.

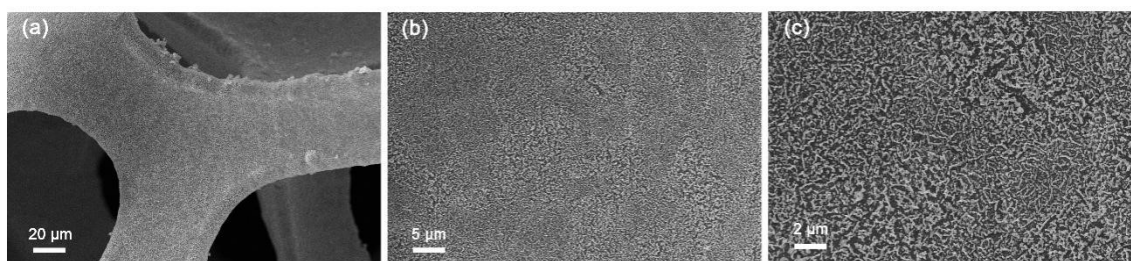


**Figure S6.** (a) The HRTEM image of the  $\text{H}_2\text{-NiMoO}_4$  nanosheet arrays; (b) the enlarge picture of (a).

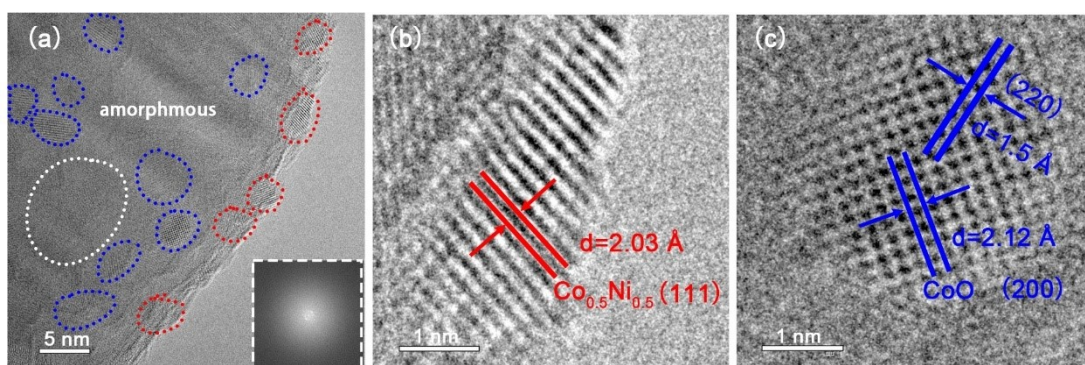




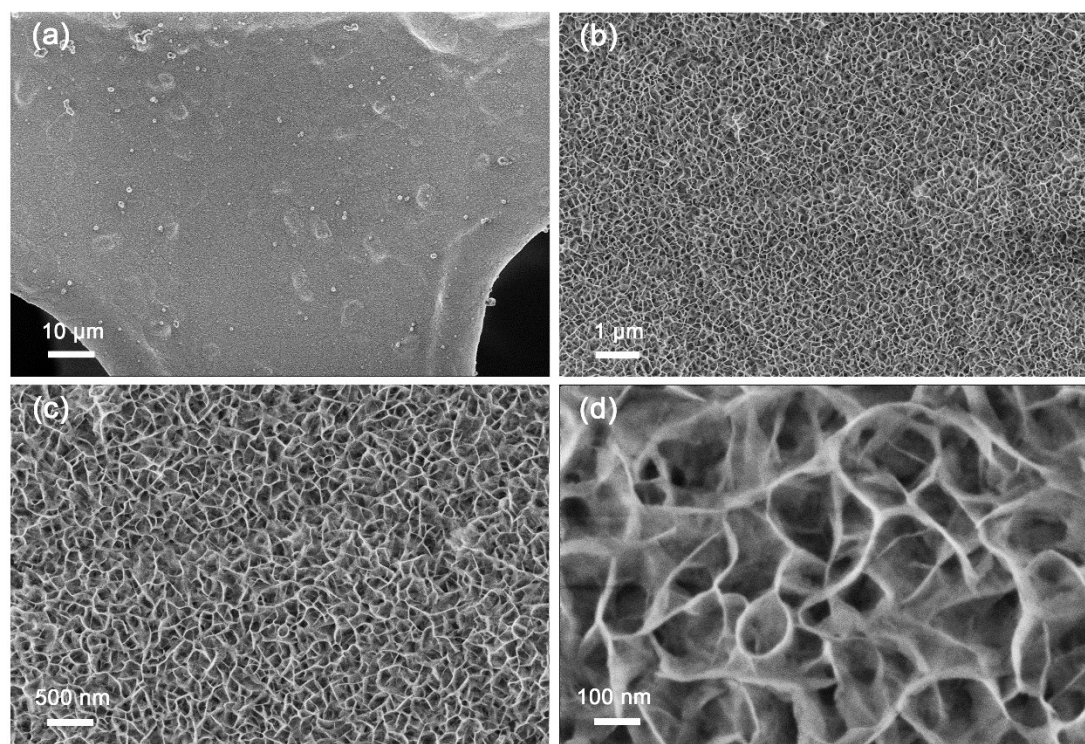
**Figure S7.** Radially integrated intensity of the diffraction patterns for  $H_2-NiMoO_4$ .



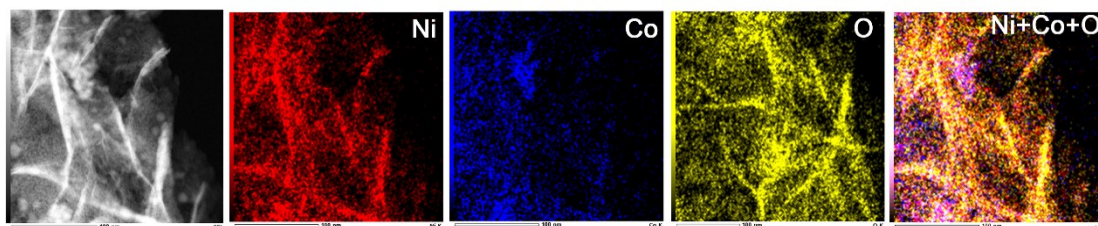
**Figure S8.** The SEM images of  $H_2-NiCo_2O_4$  on Ni foam.



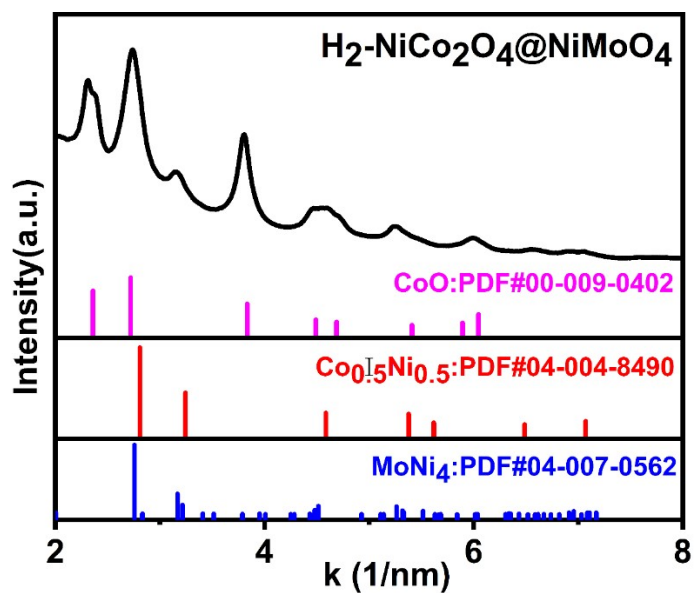
**Figure S9.** (a) The TEM image of  $\text{H}_2\text{-NiCo}_2\text{O}_4$ ; (b-d) the enlarge pictures of (a).



**Figure S10.** The SEM images of the  $\text{NiMoO}_4$  nanosheet arrays on the Ni foam.

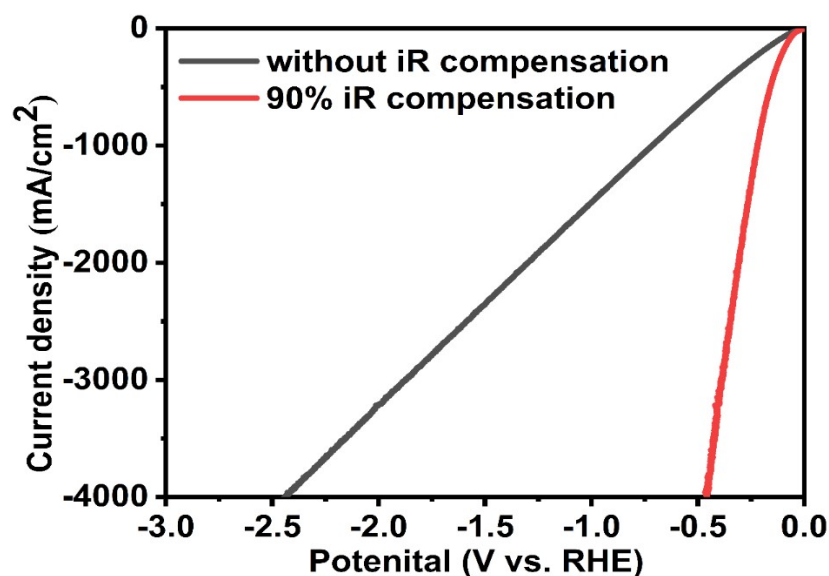


**Figure S11.** The HAADF-STEM image of  $\text{H}_2\text{-NiCo}_2\text{O}_4@\text{NiMoO}_4$  and the corresponding energy-dispersive X-ray spectroscopy (EDS) elemental dispersion mappings for Ni, Co and Mo.

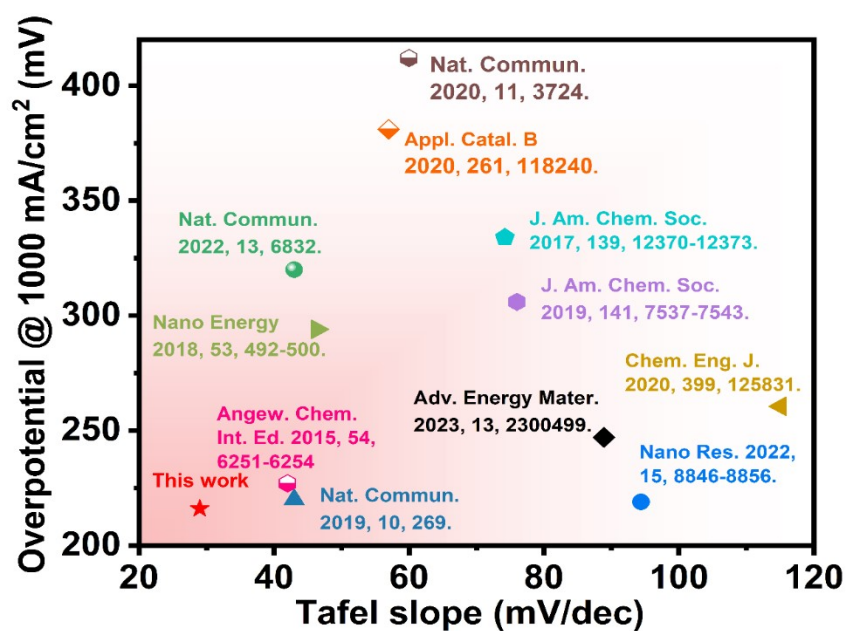


**Figure S12.** Radially integrated intensity of the diffraction patterns.



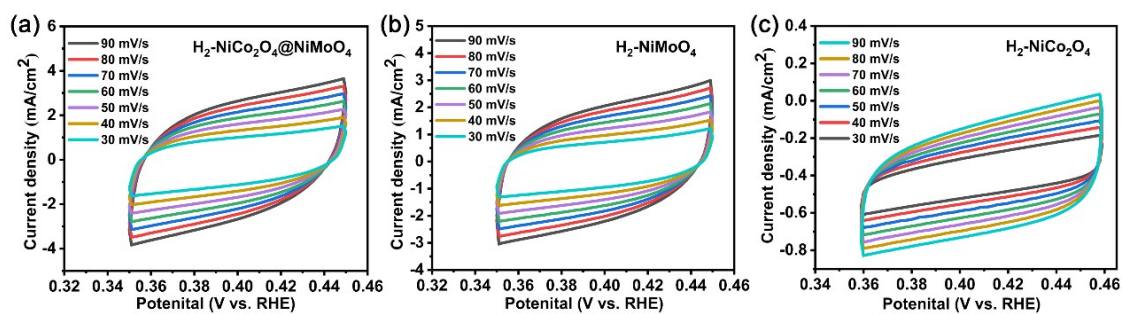


**Figure S13.** The polarization curves of  $\text{H}_2\text{-NiCo}_2\text{O}_4\text{@NiMoO}_4$  (1 M KOH) for HER without and with 90% iR compensation, where the electrolyte resistance ( $R$ ) is approximately  $1.75 \, \Omega$ .

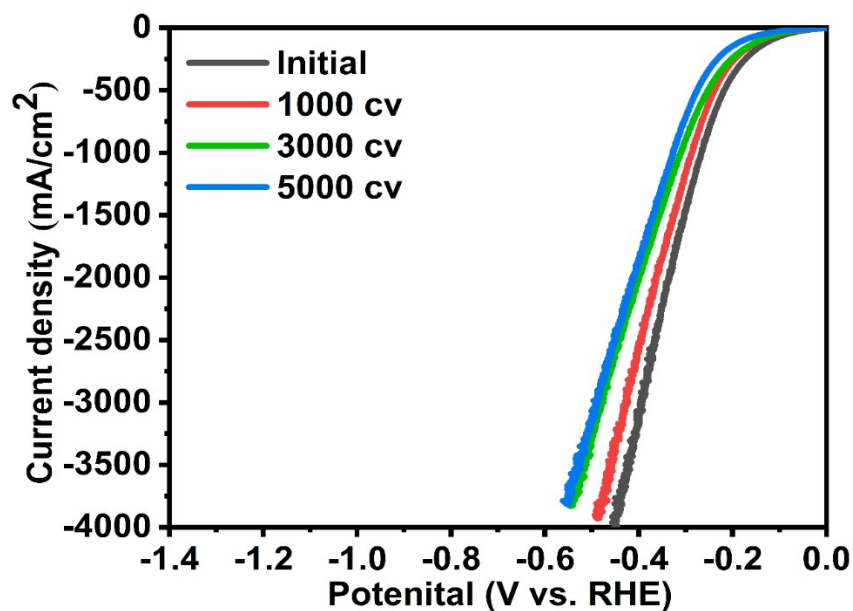


**Figure S14.** The comparative overpotentials of reported electrocatalysts for HER at  $1000 \, \text{mA/cm}^2$  in 1 M KOH electrolyte.

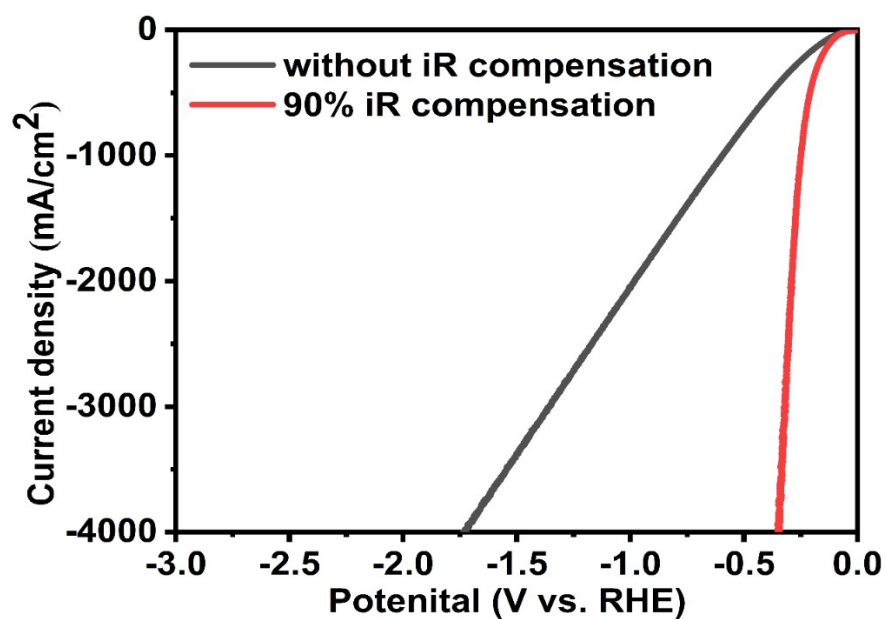




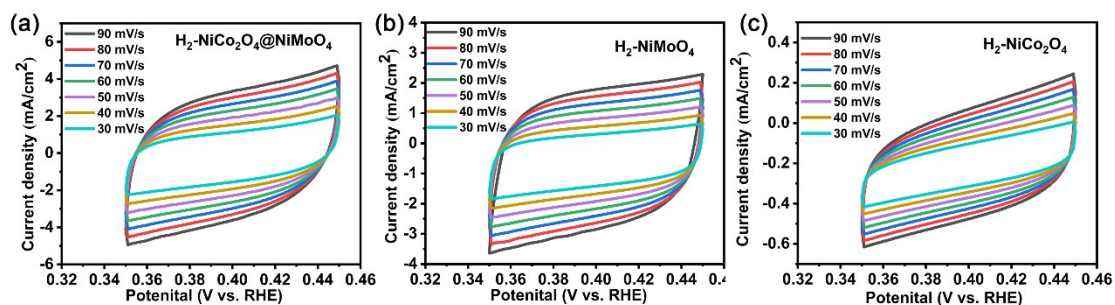
**Figure S15.** (a-c) Cyclic voltammetry curves at various scan rates of  $\text{H}_2\text{-NiCo}_2\text{O}_4@\text{NiMoO}_4$ ,  $\text{H}_2\text{-NiMoO}_4$  and  $\text{H}_2\text{-NiCo}_2\text{O}_4$  in 1.0 M KOH solution, respectively.



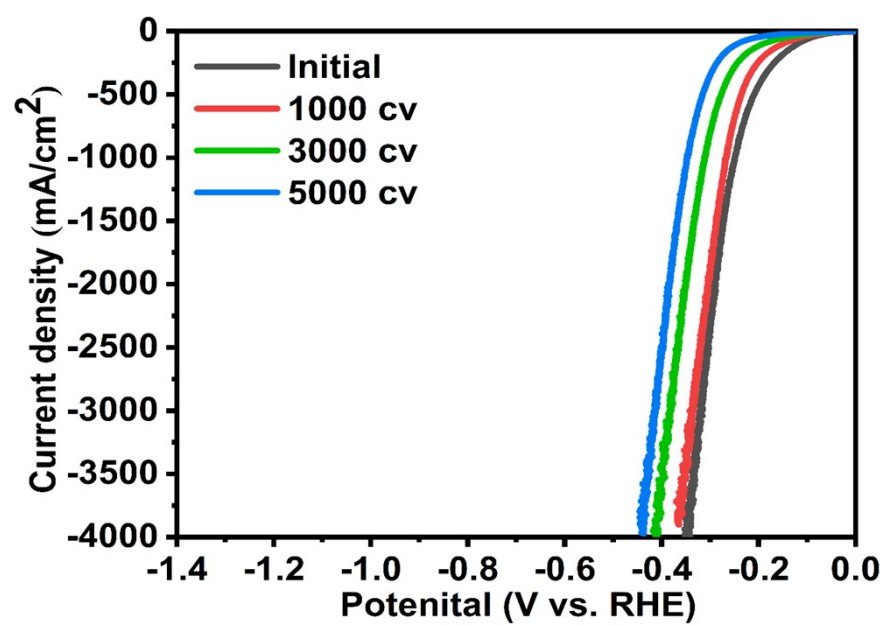
**Figure S16.** The CV stability tests of  $\text{H}_2\text{-NiCo}_2\text{O}_4@\text{NiMoO}_4$  initial and after 1000, 3000, 5000 cycles in 1.0 M KOH solution.



**Figure S17.** The polarization curves of  $\text{H}_2\text{-NiCo}_2\text{O}_4@\text{NiMoO}_4$  (1 M KOH seawater) for HER without and with 90% iR compensation.



**Figure S18.** (a-c) Cyclic voltammetry curves of  $\text{H}_2\text{-NiCo}_2\text{O}_4@\text{NiMoO}_4$ ,  $\text{H}_2\text{-NiMoO}_4$  and  $\text{H}_2\text{-NiCo}_2\text{O}_4$  in 1.0 M KOH seawater, respectively.



**Figure S19.** The CV stability tests of  $\text{H}_2\text{-NiCo}_2\text{O}_4@\text{NiMoO}_4$  initial and after 1000, 3000, 5000 cycles.



**Table S1.** The HER performance comparisons of the H<sub>2</sub>-NiCo<sub>2</sub>O<sub>4</sub>@NiMoO<sub>4</sub> with reported electrocatalysts.

| Electrocatalyst                                                      | Electrolyte | Overpotential<br>at 1000 mA/cm <sup>2</sup> | Tafel slope   | Reference |
|----------------------------------------------------------------------|-------------|---------------------------------------------|---------------|-----------|
| MoNi@NF                                                              | 1 M KOH     | 219 mV                                      | 94.5 mV/dec   | 1         |
| MoS <sub>2</sub> /Mo <sub>2</sub> C                                  | 1 M KOH     | 220 mV                                      | 43 mV/dec     | 2         |
| Co-P                                                                 | 1 M KOH     | 227 mV                                      | 42 mV/dec     | 3         |
| NiCoP-120                                                            | 1 M KOH     | 247 mV                                      | 88.9 mV/dec   | 4         |
| F-Co <sub>2</sub> P/Fe <sub>2</sub> P/IF                             | 1 M KOH     | 260.5 mV                                    | 115.01 mV/dec | 5         |
| Ni <sub>2(1-x)</sub> Mo <sub>2x</sub> P                              | 1 M KOH     | 294 mV                                      | 46.4 mV/dec   | 6         |
| Ni <sub>2</sub> P/NF                                                 | 1 M KOH     | 306 mV                                      | 76 mV/dec     | 7         |
| CuMo <sub>6</sub> S <sub>8</sub> /Cu                                 | 1 M KOH     | 320 mV                                      | 43 mV/dec     | 8         |
| $\alpha$ -MoB <sub>2</sub>                                           | 1 M KOH     | 334 mV                                      | 74.2 mV/dec   | 9         |
| A-NiCo LDH/NF                                                        | 1 M KOH     | 381 mV                                      | 57 mV/dec     | 10        |
| HC-MoS <sub>2</sub> /Mo <sub>2</sub> C                               | 1 M KOH     | 412 mV                                      | 60 mV/dec     | 11        |
| H <sub>2</sub> -NiCo <sub>2</sub> O <sub>4</sub> @NiMoO <sub>4</sub> | 1 M KOH     | 216 mV                                      | 29 mV/dec     | This work |

## References

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