

**Supporting Information**

**Synergistic effect between 1D Co<sub>3</sub>S<sub>4</sub>/MoS<sub>2</sub> heterostructures to boost  
the performance for alkaline overall water splitting**

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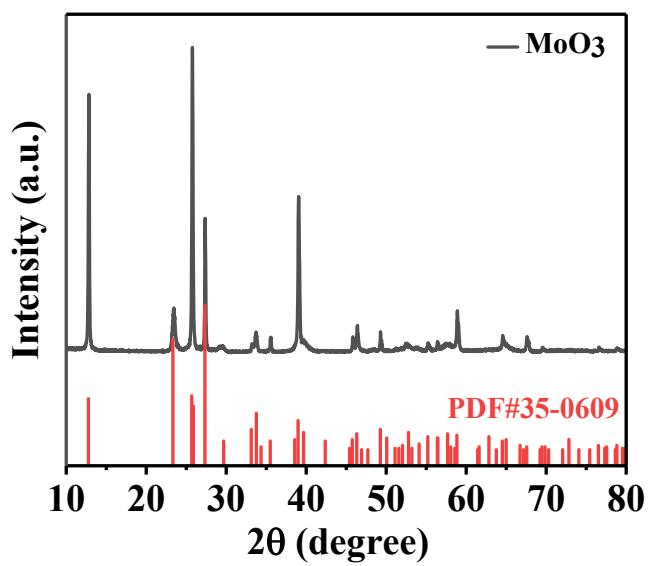
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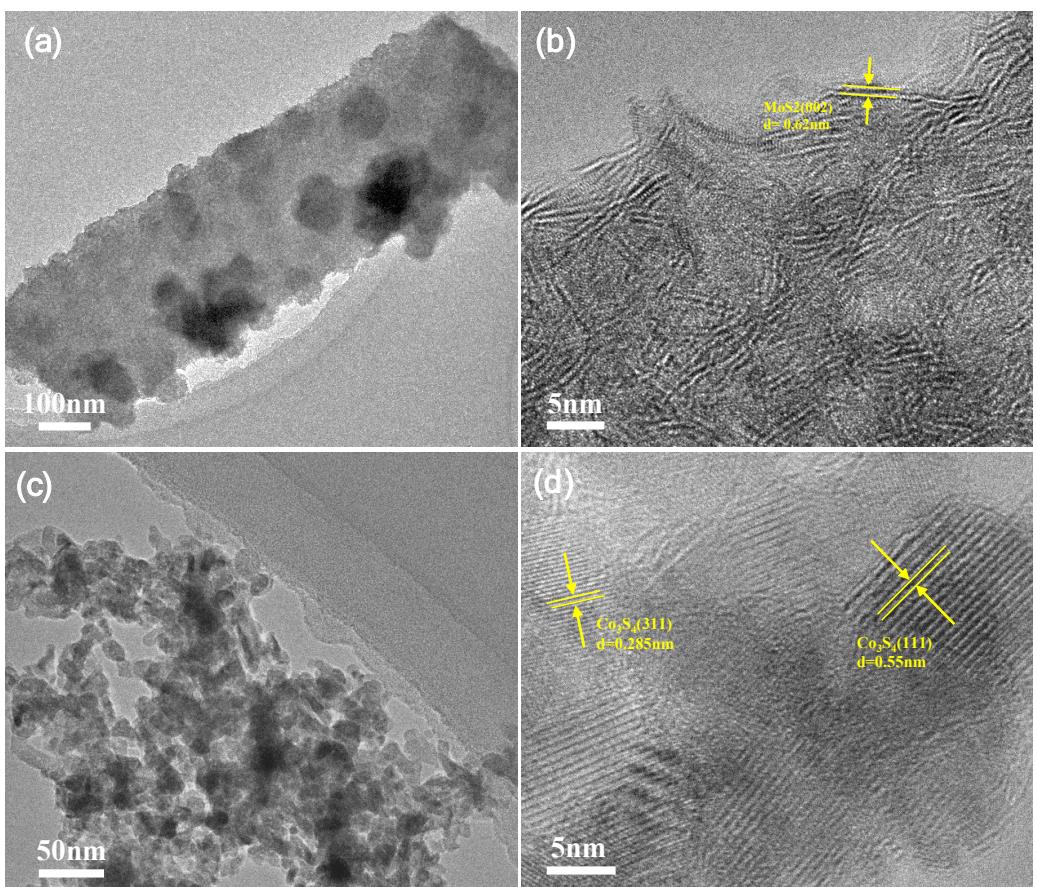
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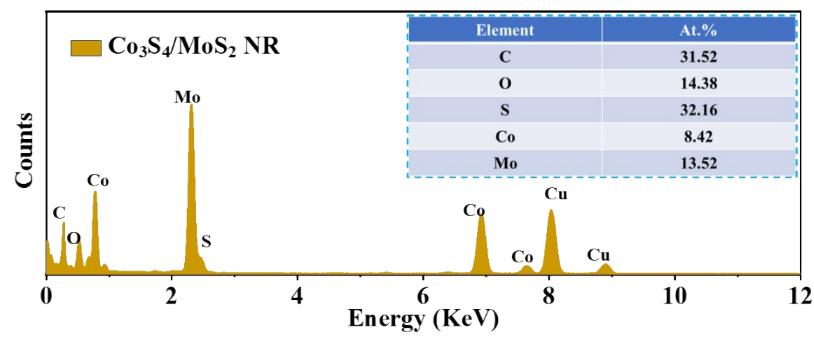
- 14 figures and 1 table



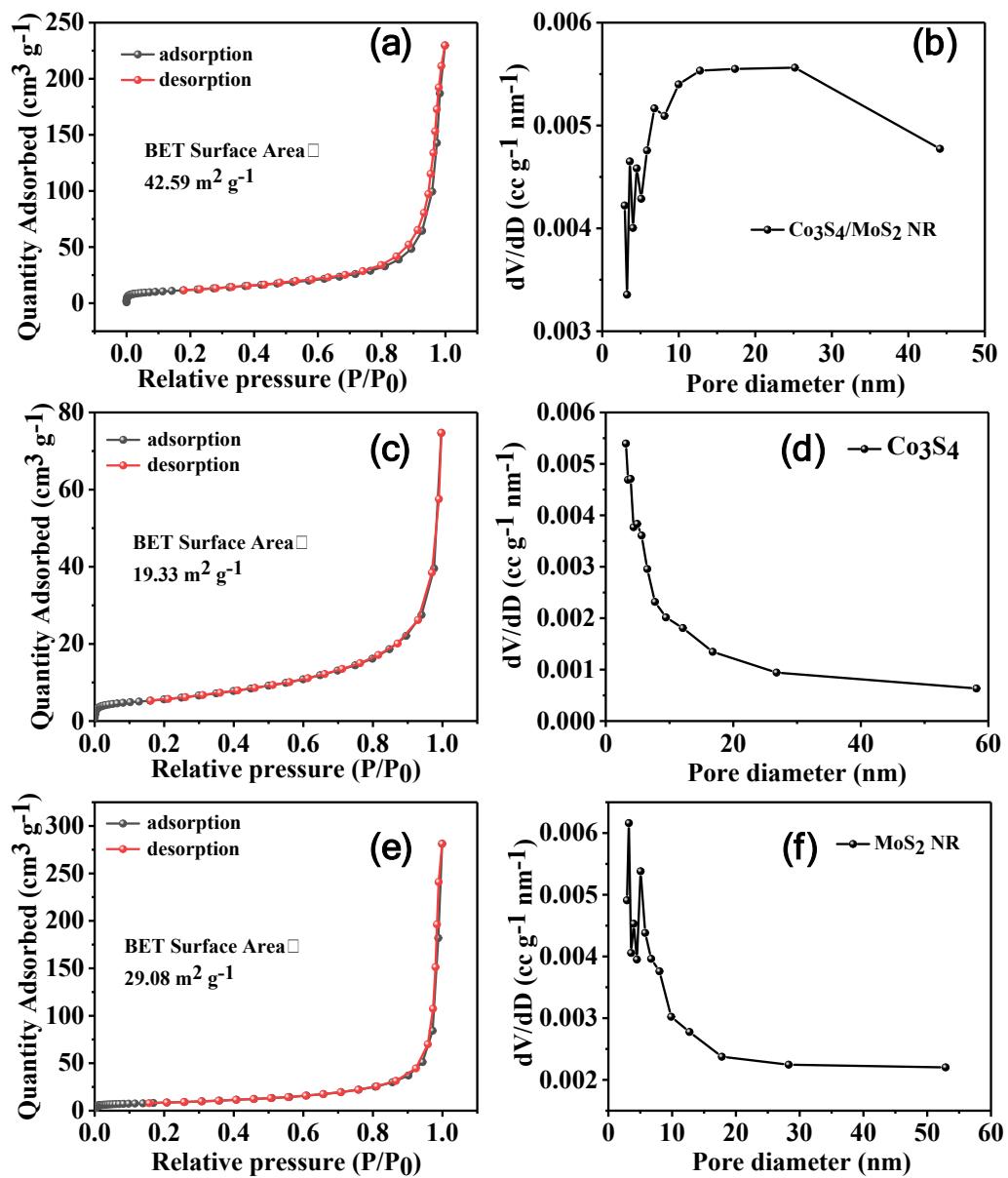
**Figure S1.** XRD pattern of  $\text{MoO}_3$ .



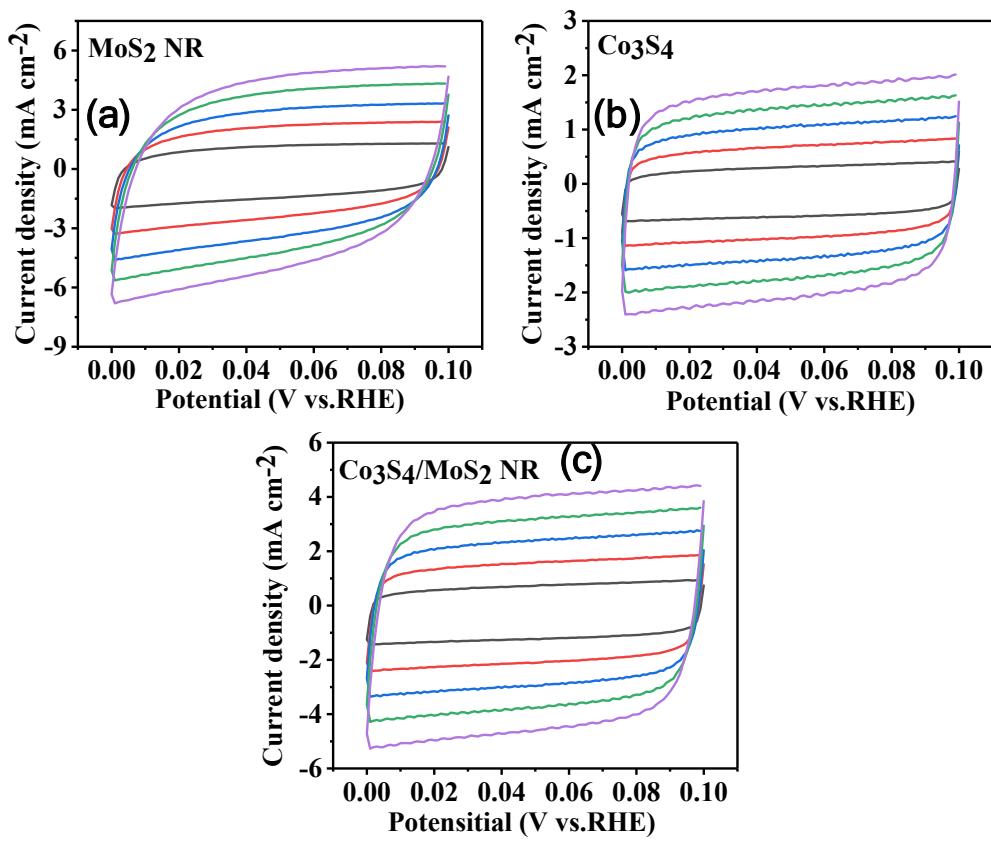
**Figure S2.** (a, c) TEM images of MoS<sub>2</sub> NR and Co<sub>3</sub>S<sub>4</sub>. (b, d) the corresponding HRTEM images of MoS<sub>2</sub> NR and Co<sub>3</sub>S<sub>4</sub>.



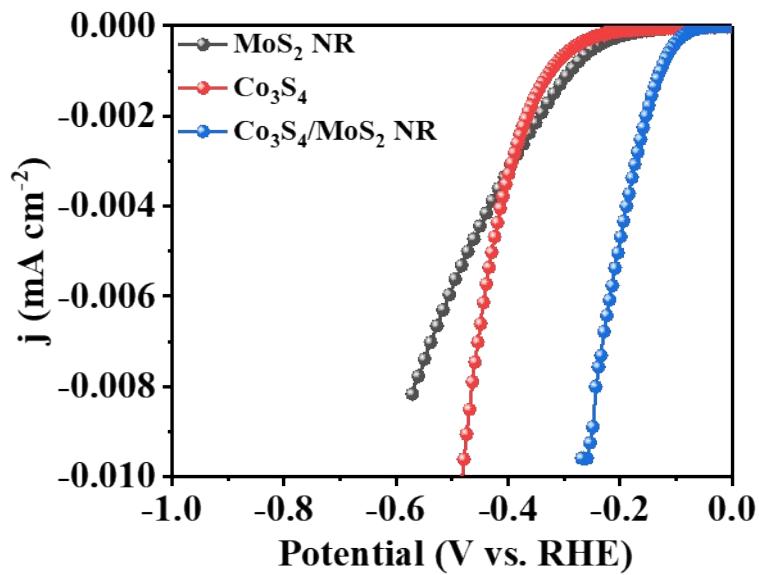
**Figure S3.** The TEM-EDS spectrum of  $\text{Co}_3\text{S}_4/\text{MoS}_2$  NR.



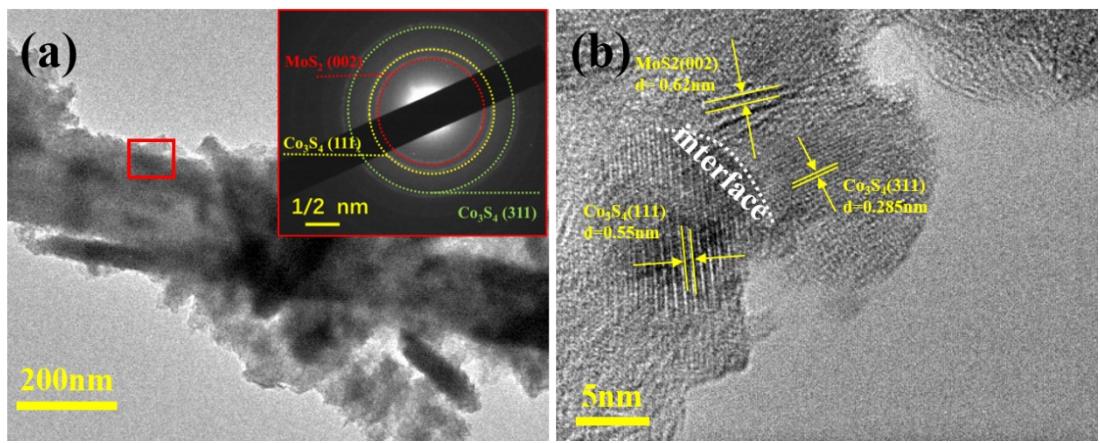
**Figure S4.** (a, c, e) Nitrogen adsorption-desorption isotherms of as-made samples. (b, d, f) the corresponding pore-size distribution plots of as-made samples.



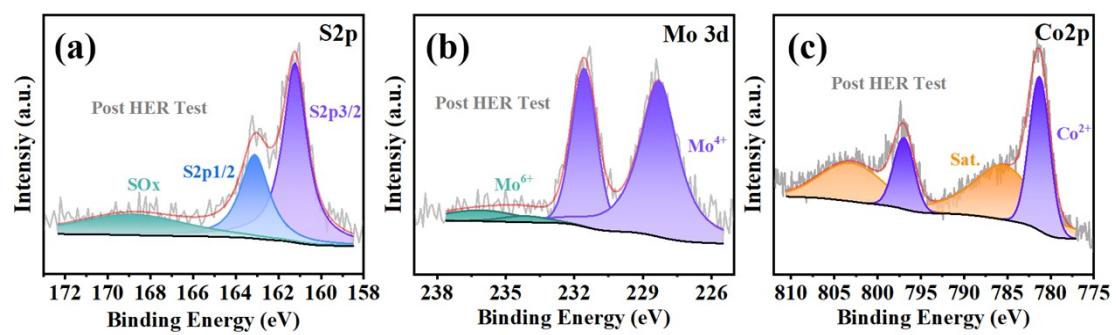
**Figure S5.** (a, b, c) CV measurements with different scan rates (40, 80, 120, 160 and 200  $\text{mV s}^{-1}$ ) at different potential ranges (0.00-0.10 V vs. RHE) in 1 M KOH solution.



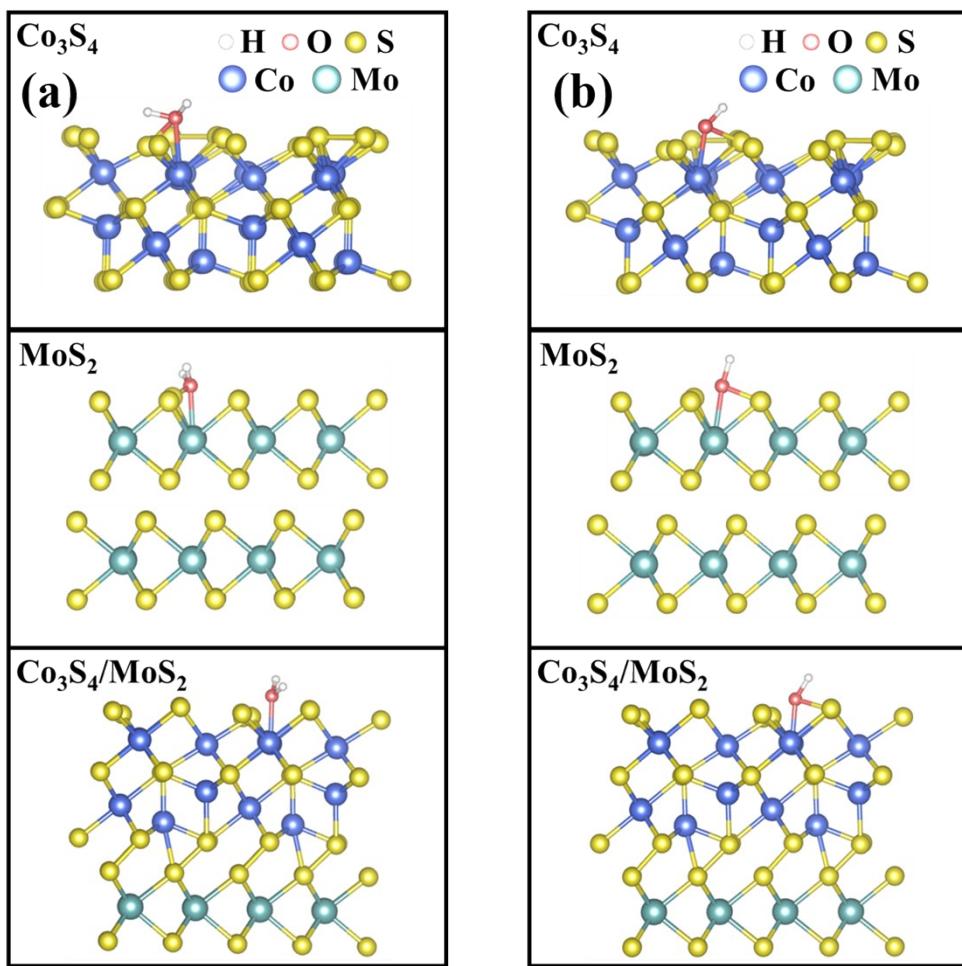
**Figure S6.** The LSV normalized by ECSA of different samples



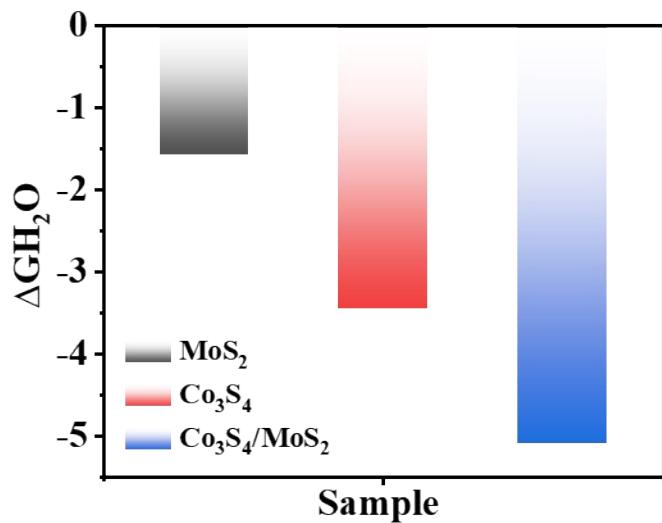
**Figure S7.** (a, b) The TEM and HRTEM images of Co<sub>3</sub>S<sub>4</sub>/MoS<sub>2</sub> NR after long-time HER test.



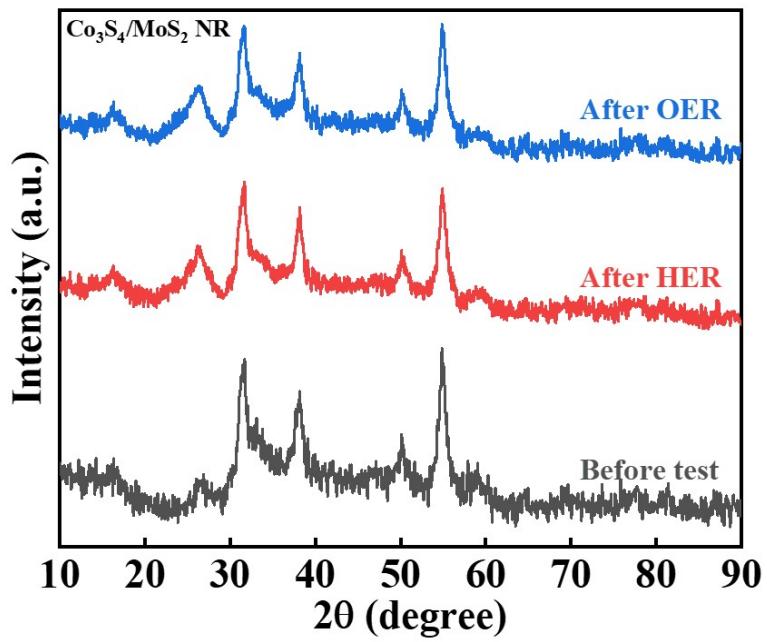
**Figure S8.** (a, b, c) The S 2p, Mo 3d and Co 2p of  $\text{Co}_3\text{S}_4/\text{MoS}_2$  NR after long-time HER test.



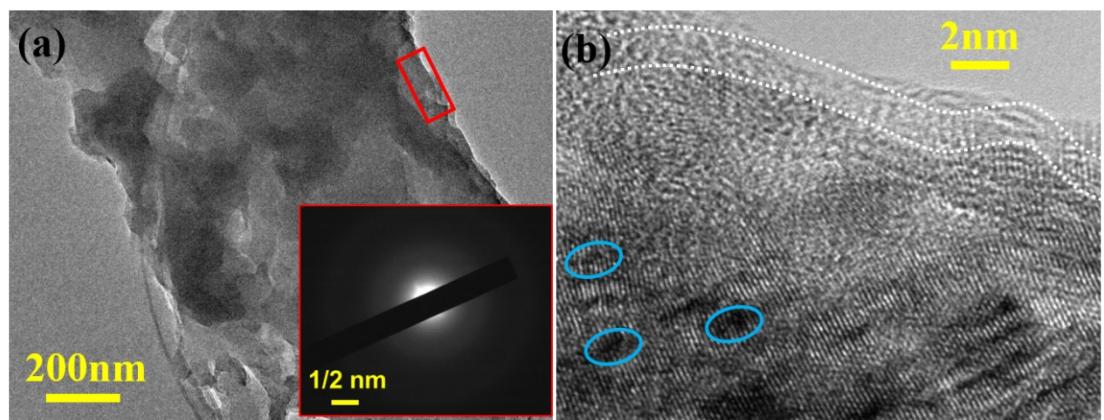
**Figure S9.** Scheme of  $\text{Co}_3\text{S}_4$  (111),  $\text{MoS}_2$  (002) and  $\text{Co}_3\text{S}_4/\text{MoS}_2$  heterostructure with  $\text{H}_2\text{O}$  (a) and  $\text{OH}$  (b) adsorbed on the respective surface.



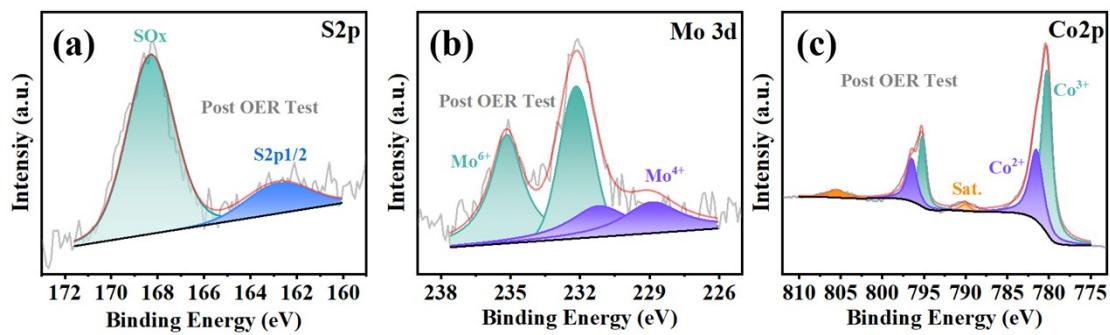
**Figure S10.** DFT-calculated  $\text{H}_2\text{O}$  free-energy change of  $\text{Co}_3\text{S}_4$  (111),  $\text{MoS}_2$  (002) and  $\text{Co}_3\text{S}_4/\text{CoS}_2$  heterostructure.



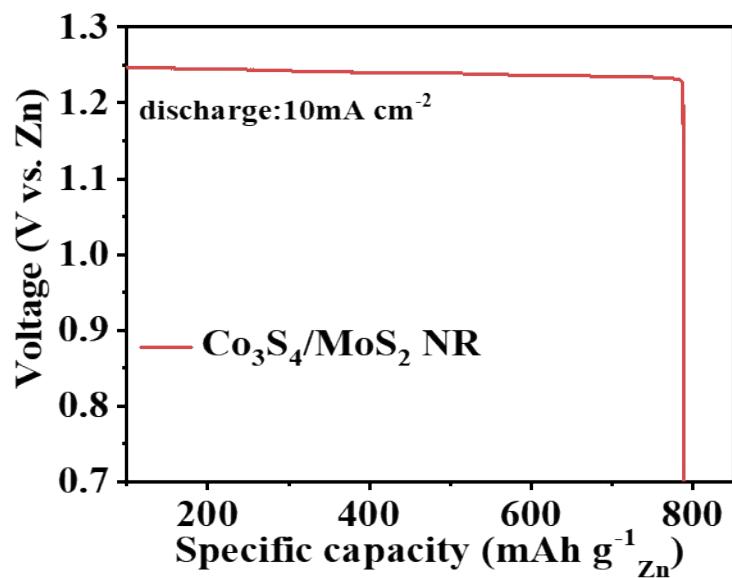
**Figure S11.** XRD pattern of  $\text{Co}_3\text{S}_4/\text{MoS}_2$  NR before and after HER and OER test.



**Figure S12.** (a, b) The TEM and HRTEM images of  $\text{Co}_3\text{S}_4/\text{MoS}_2$  NR after long-time OER test.



**Figure S13.** (a, b, c) The S 2p, Mo 3d and Co 2p of  $\text{Co}_3\text{S}_4/\text{MoS}_2$  NR after long-time OER test.



**Figure S14.** The galvanostatic discharge curves of a r-ZAB loaded with  $\text{Co}_3\text{S}_4/\text{MoS}_2$  NR catalysts on air cathode

**Table S1.** Comparison of the electrochemical HER and OER performance of  $\text{Co}_3\text{S}_4/\text{MoS}_2$  NR with some leading non-precious bifunctional electrocatalysts.

Electrocatalyst	Catalytic performance		Reference information
	Overpotential ( $j_{10}$ ) of HER (mV)	Overpotential ( $j_{10}$ ) of OER (mV)	
<b><math>\text{Co}_3\text{S}_4/\text{MoS}_2</math> NR</b>	<b>116</b>	<b>280</b>	<b>This work</b>
Co@Co-P@NPCNTs	160	290	Ref.44 (2020)
FeOOH/ $\text{Ni}_3\text{N}$	67	244	Ref.45 (2020)
$\text{Co}_4\text{N}@\text{NC}$	62	257	Ref.46 (2020)
VOOH-3Fe	90	195	Ref.47 (2020)
CoFeO@BP	88	266	Ref.48 (2020)
Co/CoP@HOMC	120	260	Ref.49 (2021)
WN-Ni@N,P-CNT	70	268	Ref.50 (2021)
Ni-Mo-P	69	235	Ref.51 (2021)
$\text{MoS}_2/\text{NiFe-LDH}$	110	210	Ref.52 (2019)
Co-Ni <sub>3</sub> N	194	307	Ref.53 (2018)
NiFe-MOF-5	163	168	Ref.54 (2021)
$\text{Co}_9\text{S}_8@\text{Co}_9\text{S}_8@\text{MoS}_{2-x}$	173	340	Ref.55 (2019)
Co@N-CNTF	220	350	Ref.56 (2019)
$\text{FeS}_2@\text{MXene}$	87	240	Ref.57 (2022)
Ni-Co sulfide/NF	190	230	Ref.58 (2021)