

## Supporting Information

**The preparation of Co-precursor.** In detail, 66.7mM of  $\text{Co}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$  solution was prepared as the electrolyte, the pre-treated NF was served as the cathode, and a saturated calomel electrode (SCE) and platinum were used as reference electrode and counter electrode, respectively. The electrodeposit process was proceeded for 800s under a constant potential of -0.8 V. Further, the  $\text{Co}(\text{OH})_2/\text{NF}$  was rinsed with deionized water and ethanol and then vacuumed for 10h. The as-prepared  $\text{Co}(\text{OH})_2/\text{NF}$  was heated for 2h at  $400^\circ\text{C}$  with a heating rate of  $5^\circ\text{C}/\text{min}$  and cooled down to room temperature. Finally, a certain mass of Mo-Co(OH)<sub>2</sub> nanoplates were deposited on the  $\text{Co}_3\text{O}_4/\text{NF}$  via a similar electrodeposition process.

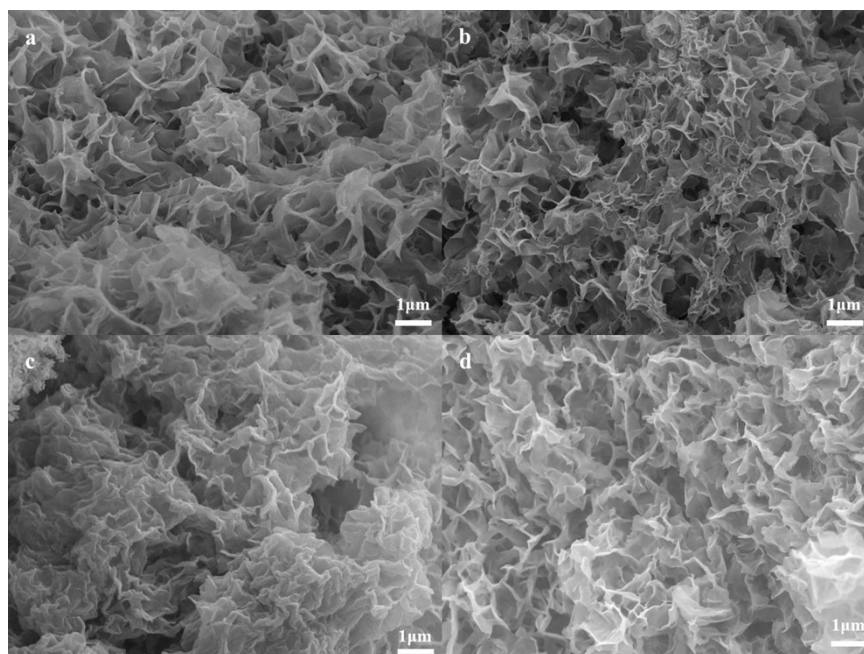


Fig. S1 SEM images of (a) Mo-CoP/Co<sub>2</sub>P/NF-300, (b) Mo-CoP/Co<sub>2</sub>P/NF-200, (c) Mo-CoP/Co<sub>2</sub>P/NF-500 and (d) Mo-CoP/Co<sub>2</sub>P/NF-800.

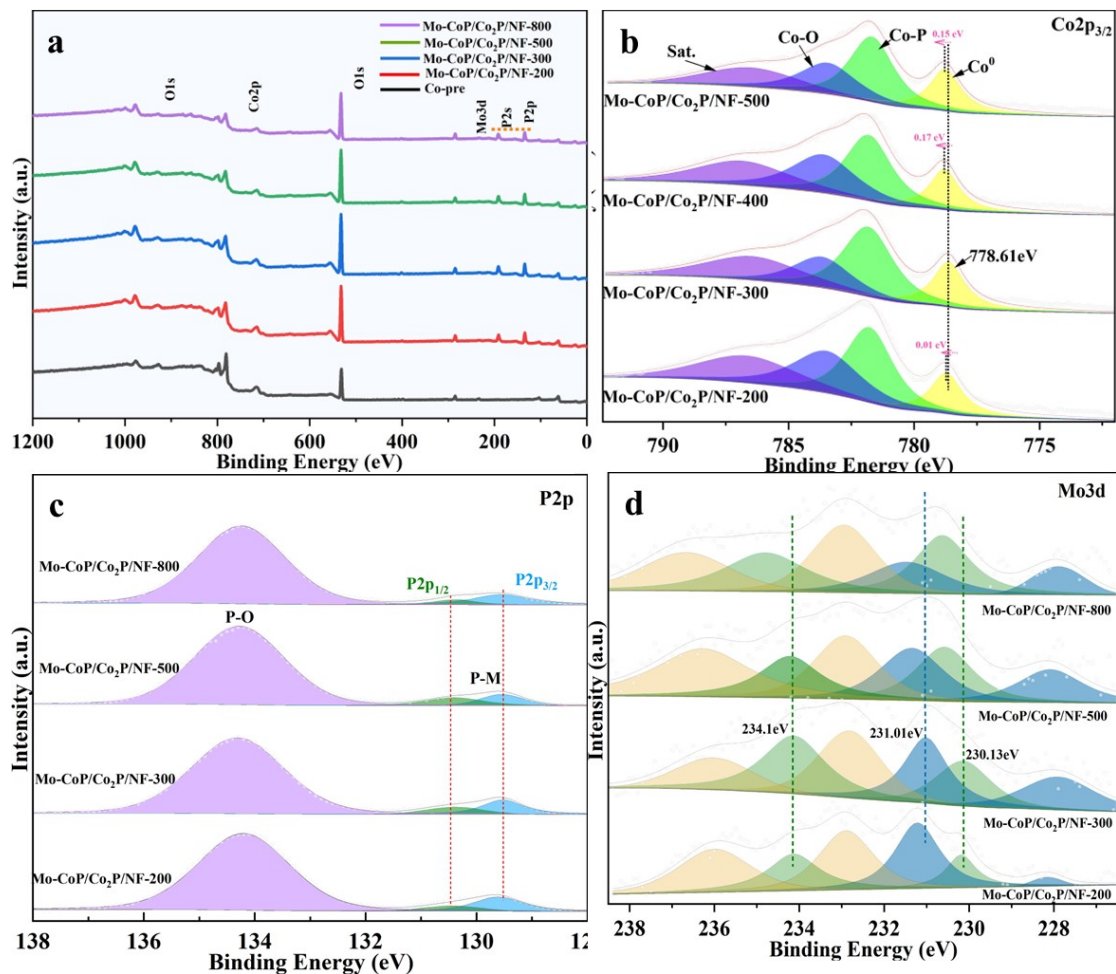


Fig. S2 (a) Survey X-ray photoelectron spectroscopy (XPS) of Mo-CoP/Co<sub>2</sub>P/NF-300, Mo-CoP/Co<sub>2</sub>P/NF-200, Mo-CoP/Co<sub>2</sub>P/NF-500 and Mo-CoP/Co<sub>2</sub>P/NF-800. (b-d) XPS spectra of Co<sub>2</sub>p, P2p and Mo3d for Mo-CoP/Co<sub>2</sub>P/NF-300, Mo-CoP/Co<sub>2</sub>P/NF-200, Mo-CoP/Co<sub>2</sub>P/NF-500 and Mo-CoP/Co<sub>2</sub>P/NF-800.

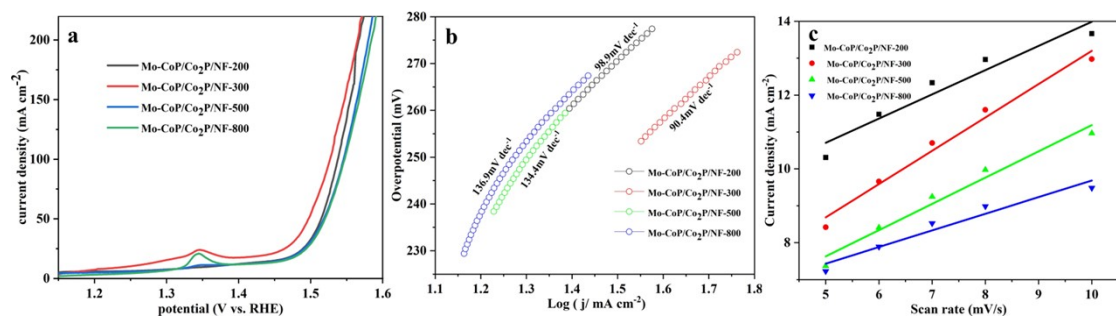


Fig. S3 (a) LSV curves, (b) Tafel slopes and (c) the double-layer capacitance (C<sub>dl</sub>) of CoP/Co<sub>2</sub>P/NF-200, CoP/Co<sub>2</sub>P/NF-300, Mo-CoP/Co<sub>2</sub>P/NF-500 and Mo-CoP/Co<sub>2</sub>P/NF-800 for HER

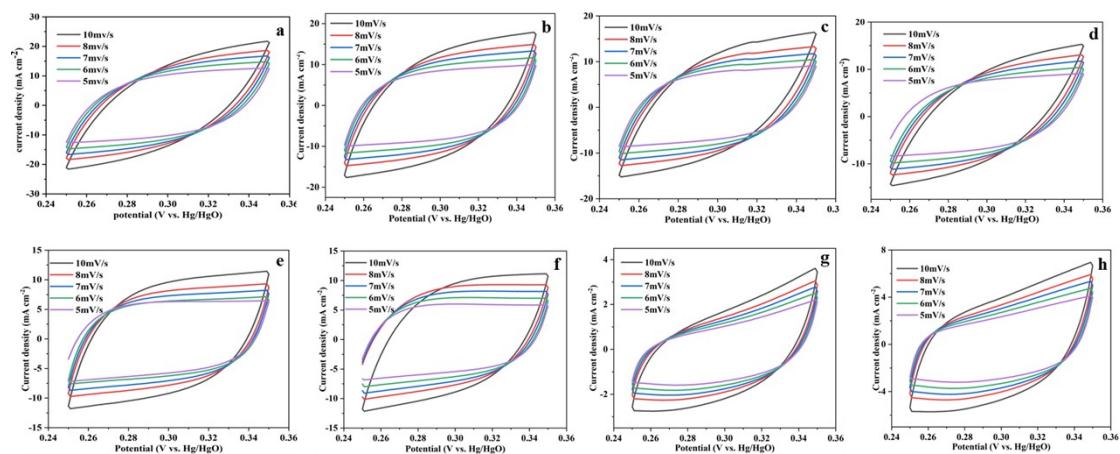


Fig. S4 CV curves of the CoP/Co<sub>2</sub>P/NF-200, CoP/Co<sub>2</sub>P/NF-300, Mo-CoP/Co<sub>2</sub>P/NF-500, Mo-CoP/Co<sub>2</sub>P/NF-800, Co-pre, CoP, Co<sub>2</sub>P, CoP/Co<sub>2</sub>P at the different scan rates ranging from 5 to 10 mV s<sup>-1</sup> for OER.

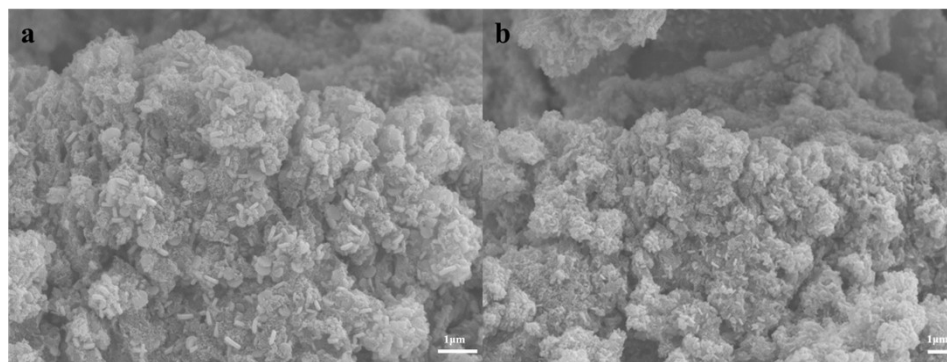


Fig. S5 SEM images of CoP/Co<sub>2</sub>P/NF-300 after OER

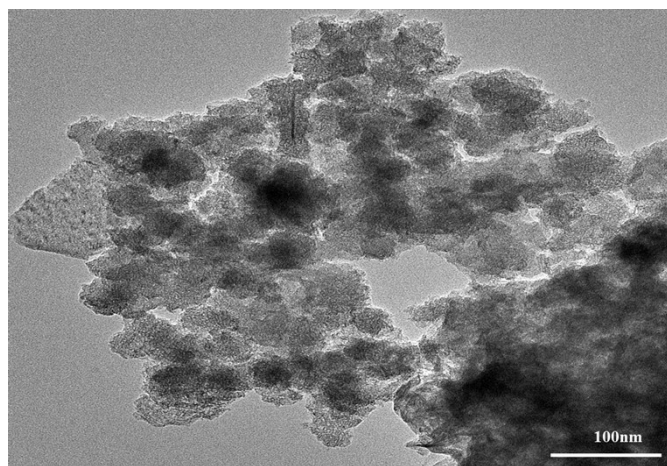


Fig. S6 TEM images of CoP/Co<sub>2</sub>P/NF-300 after OER

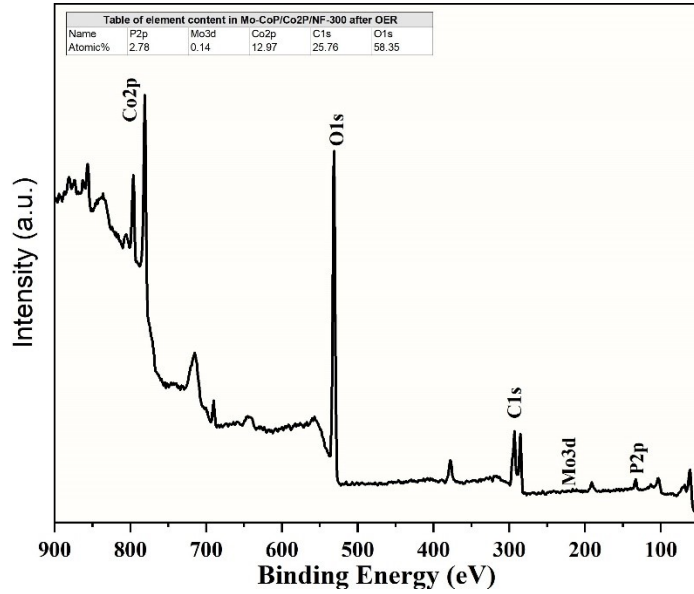


Fig. S7 Survey spectra of CoP/Co<sub>2</sub>P/NF-300 after OER

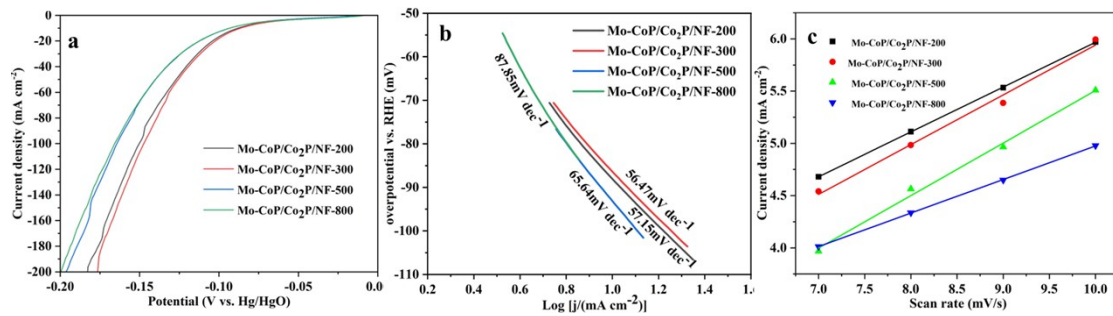


Fig. S8 (a) LSV curves, (b) Tafel slopes and (c) the double-layer capacitance (Cdl) of CoP/Co<sub>2</sub>P/NF-200, CoP/Co<sub>2</sub>P/NF-300, Mo-CoP/Co<sub>2</sub>P/NF-500 and Mo-CoP/Co<sub>2</sub>P/NF-800 for OER

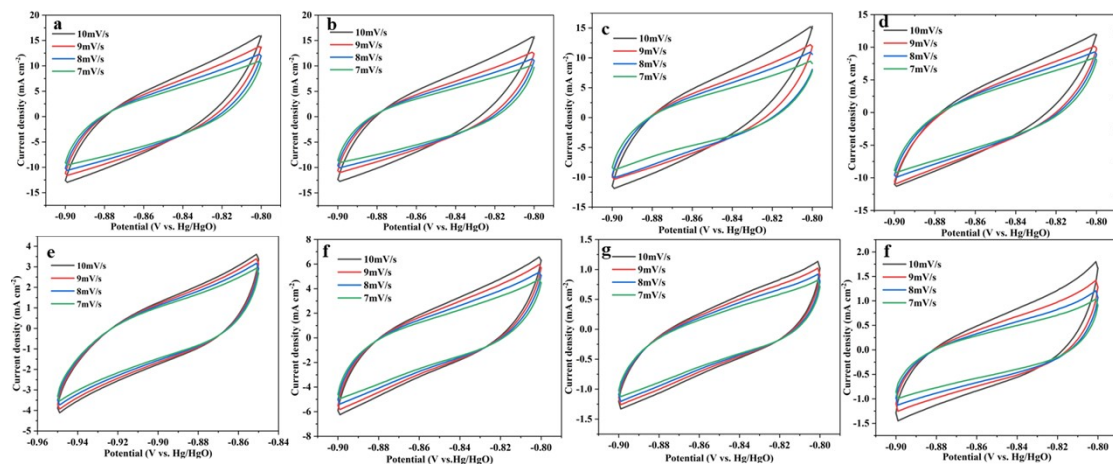


Fig. S9 CV curves of the CoP/Co<sub>2</sub>P/NF-200, CoP/Co<sub>2</sub>P/NF-300, Mo-CoP/Co<sub>2</sub>P/NF-500, Mo-CoP/Co<sub>2</sub>P/NF-800, Co-pre, CoP, Co<sub>2</sub>P, CoP/Co<sub>2</sub>P at the different scan rates ranging from 5 to 10 mV s<sup>-1</sup> for HER.

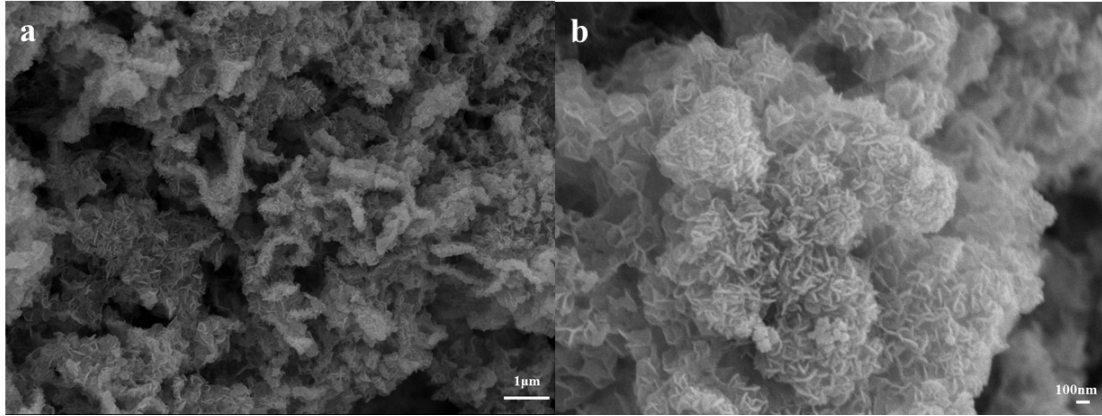


Fig. S10 SEM images of CoP/Co<sub>2</sub>P/NF-300 after HER

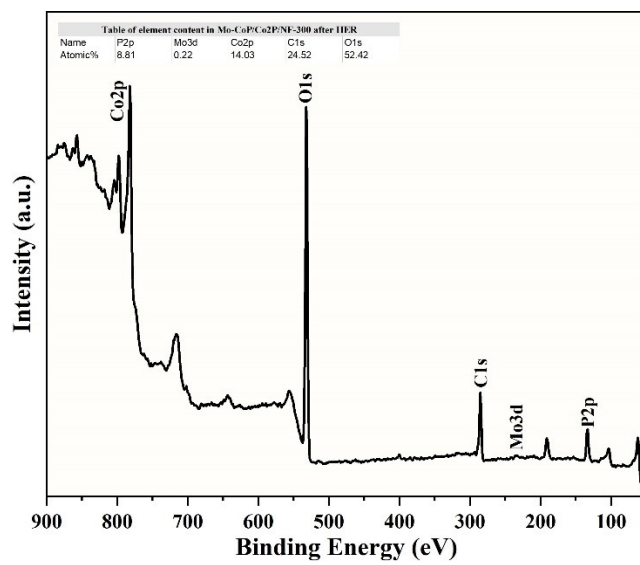


Fig. S11 Survey spectra of CoP/Co<sub>2</sub>P/NF-300 after HER

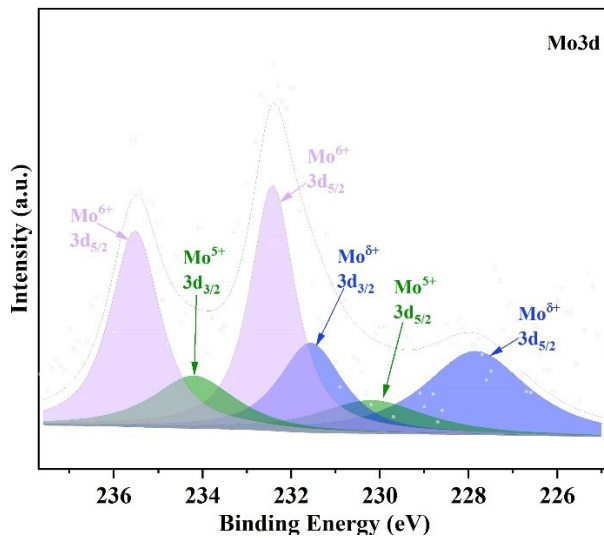


Fig. S12 High-resolution XPS spectra of Mo3d in Mo-CoP/Co<sub>2</sub>P/NF-300 after HER

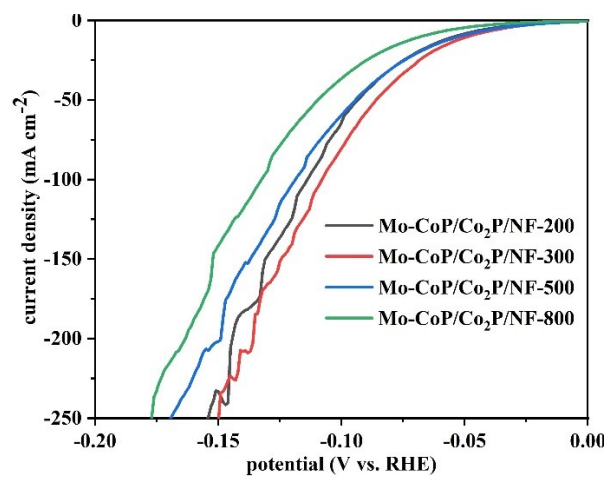


Fig. S13 LSV curves for HER in 0.5M H<sub>2</sub>SO<sub>4</sub> of Mo-CoP/Co<sub>2</sub>P/NF-200, Mo-CoP/Co<sub>2</sub>P/NF-300, Mo-CoP/Co<sub>2</sub>P/NF-500 and Mo-CoP/Co<sub>2</sub>P/NF-800

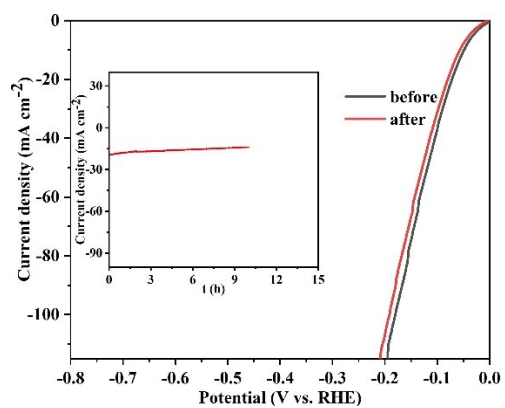


Fig. S14 Mo-CoP/Co<sub>2</sub>P/NF-300 polarization curves of before and after 10h for HER in H<sub>2</sub>SO<sub>4</sub>. The inset gives the chronoamperometric curve of the HER for 10h.