Supporting Information

The preparation of Co-precursor. In detail, 66.7mM of Co(NO$_3$)$_2$•6H$_2$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 electrodeposition process was proceeded for 800s under a constant potential of -0.8 V. Further, the Co(OH)$_2$/NF was rinsed with deionized water and ethanol and then vacuumed for 10h. The as-prepared Co(OH)$_2$/NF was heated for 2h at 400℃ with a heating rate of 5℃/min and cooled down to room temperature. Finally, a certain mass of Mo-Co(OH)$_2$ nanoplates were deposited on the Co$_3$O$_4$/NF via a similar electrodeposition process.

Fig. S1 SEM images of (a) Mo-CoP/Co$_2$P/NF-300, (b) Mo-CoP/Co$_2$P/NF-200, (c) Mo-CoP/Co$_2$P/NF-500 and (d) Mo-CoP/Co$_2$P/NF-800.
Fig. S2 (a) Survey X-ray photoelectron spectroscopy (XPS) of Mo-CoP/Co$_2$P/NF-300, Mo-CoP/Co$_2$P/NF-200, Mo-CoP/Co$_2$P/NF-500 and Mo-CoP/Co$_2$P/NF-800. (b-d) XPS spectra of Co2p, P2p and Mo3d for Mo-CoP/Co$_2$P/NF-300, Mo-CoP/Co$_2$P/NF-200, Mo-CoP/Co$_2$P/NF-500 and Mo-CoP/Co$_2$P/NF-800.

Fig. S3 (a) LSV curves, (b) Tafel slopes and (c) the double-layer capacitance (Cdl) of CoP/Co$_2$P/NF-200, CoP/Co$_2$P/NF-300, Mo-CoP/Co$_2$P/NF-500 and Mo-CoP/Co$_2$P/NF-800 for HER.
Fig. S4 CV curves of the CoP/Co$_2$P/NF-200, CoP/Co$_2$P/NF-300, Mo-CoP/Co$_2$P/NF-500, Mo-CoP/Co$_2$P/NF-800, Co-pre, CoP, Co$_2$P, CoP/Co$_2$P at the different scan rates ranging from 5 to 10 mV s$^{-1}$ for OER.

Fig. S5 SEM images of CoP/Co$_2$P/NF-300 after OER

Fig. S6 TEM images of CoP/Co$_2$P/NF-300 after OER
Fig. S7 Survey spectra of CoP/Co\textsubscript{2}P/NF-300 after OER

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

Fig. S9 CV curves of the CoP/Co\textsubscript{2}P/NF-200, CoP/Co\textsubscript{2}P/NF-300, Mo-CoP/Co\textsubscript{2}P/NF-500, Mo-CoP/Co\textsubscript{2}P/NF-800, Co-pre, CoP, Co\textsubscript{3}P, CoP/Co\textsubscript{2}P at the different scan rates ranging from 5 to 10 mV s\textsuperscript{-1} for HER.
Fig. S10 SEM images of CoP/Co$_2$P/NF-300 after HER

![SEM images of CoP/Co$_2$P/NF-300 after HER](image)

Fig. S11 Survey spectra of CoP/Co$_2$P/NF-300 after HER

![Survey spectra of CoP/Co$_2$P/NF-300 after HER](image)

Fig. S12 High-resolution XPS spectra of Mo3d in Mo-CoP/Co$_2$P/NF-300 after HER

![High-resolution XPS spectra of Mo3d in Mo-CoP/Co$_2$P/NF-300 after HER](image)
Fig. S13 LSV curves for HER in 0.5M H2SO4 of Mo-CoP/Co3P/NF-200, Mo-CoP/Co3P/NF-300, Mo-CoP/Co3P/NF-500 and Mo-CoP/Co3P/NF-800.

Fig. S14 Mo-CoP/Co3P/NF-300 polarization curves of before and after 10h for HER in H2SO4. The inset gives the chronoamperometric curve of the HER for 10h.