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Supporting Information

Interfacial engineering of nickel/iron/ruthenium phosphides for efficient overall water splitting powered by solar energy

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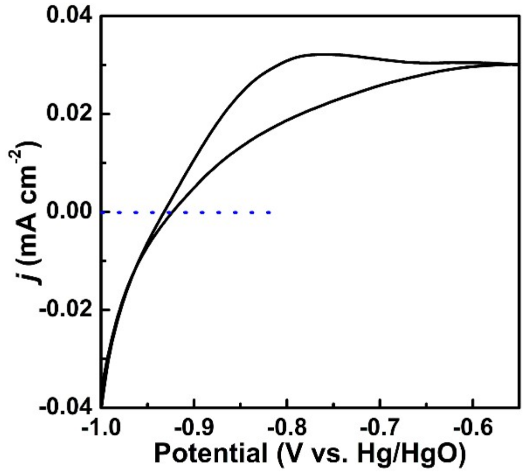

Fig. S1 Single cycle CV curves of Hg/HgO electrode calibration in 1.0 M KOH.

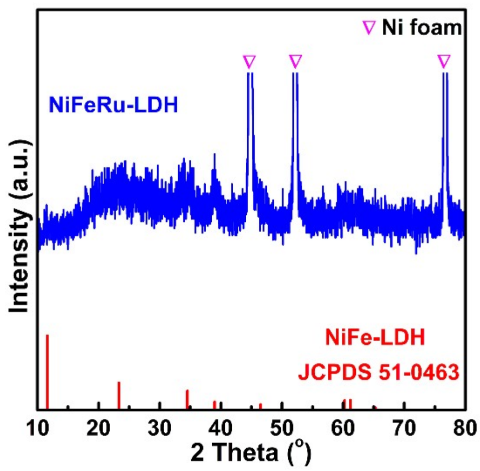

Fig. S2 PXRD patterns of NiFeRu-LDH.

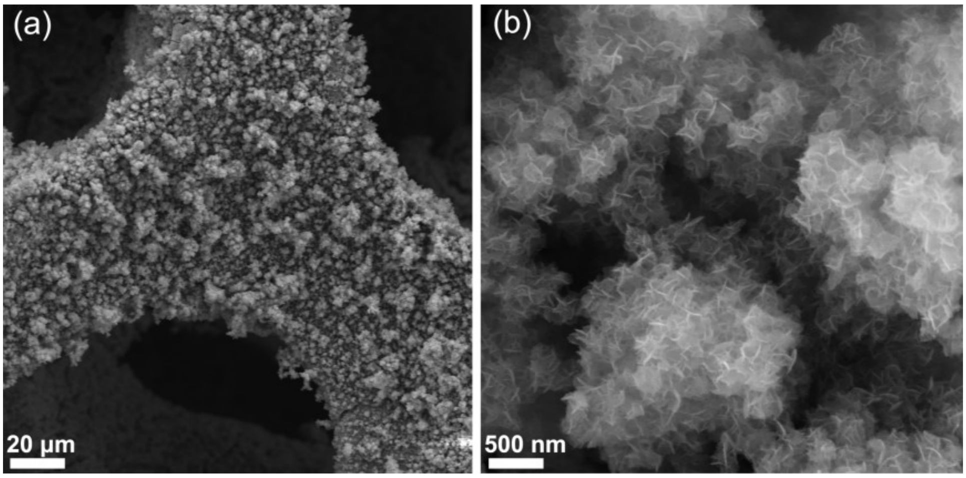

Fig. S3 (a, b) SEM images of NiFeRu-LDH/NF.

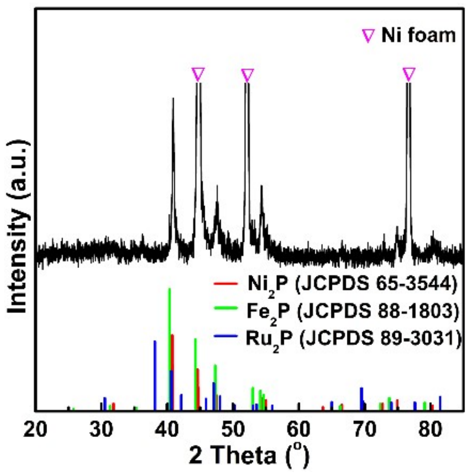

Fig. S4 PXRD patterns of Ni_2P-Fe_2P-Ru_2P/NF.

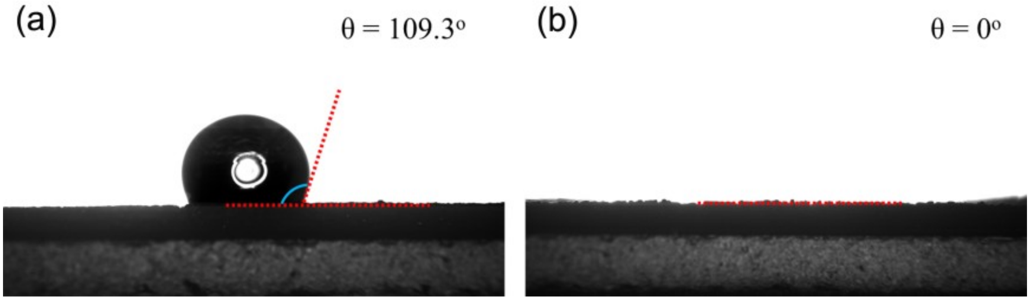

Fig. S5 Contact angles of NF (a) and Ni_2P-Fe_2P-Ru_2P /NF (b).

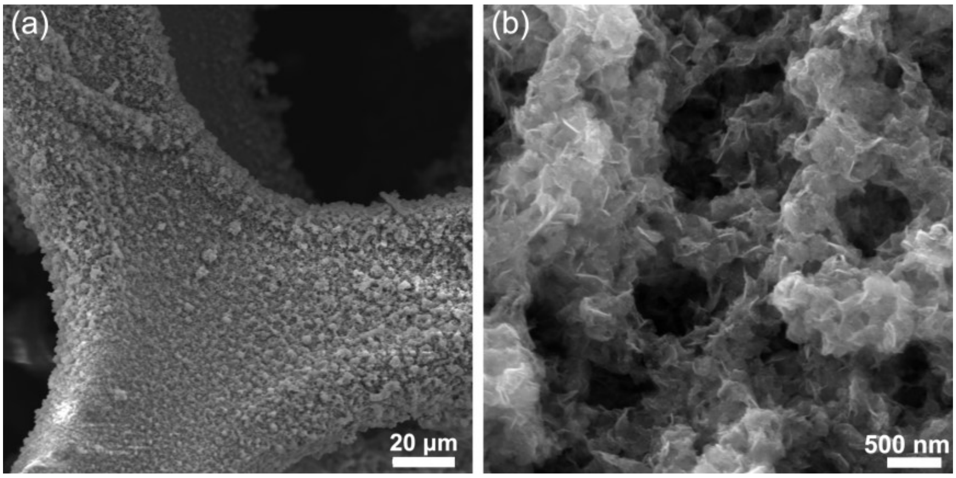

Fig. S6 (a, b) SEM images of NiFe-LDH/NF.

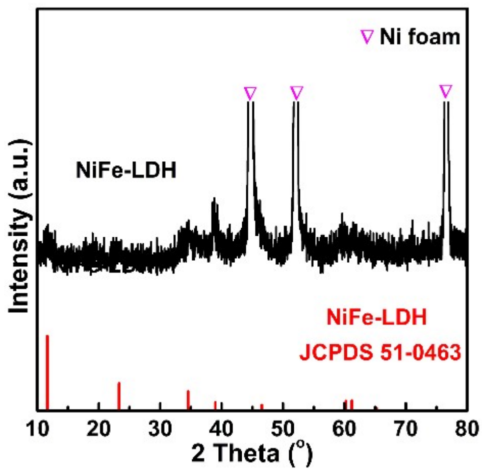

Fig. S7 PXRD patterns of NiFe-LDH.

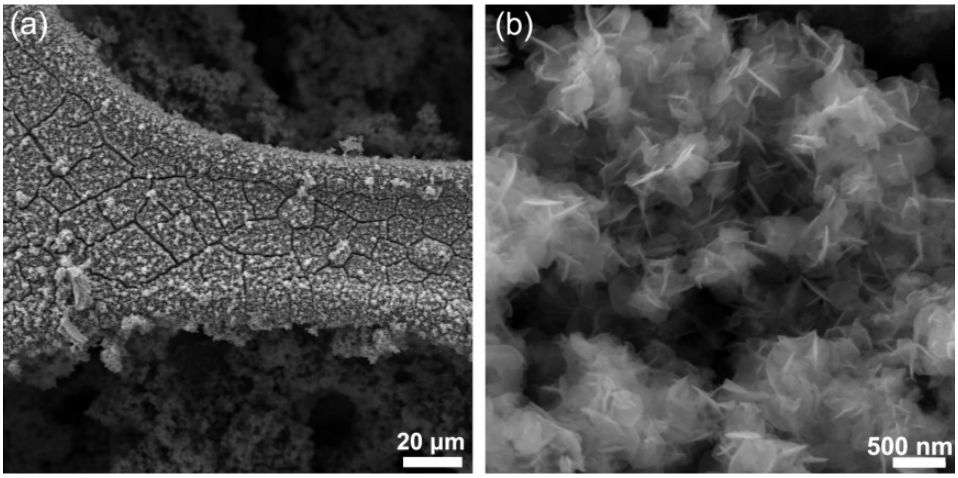

Fig. S8 (a, b) SEM images of Ni_2P-Fe_2P/NF.

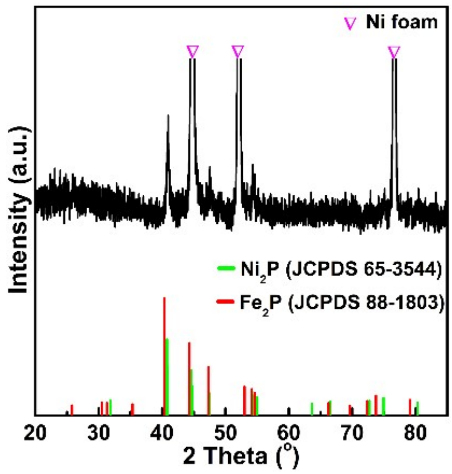

Fig. S9 PXRD patterns of Ni_2P-Fe_2P/NF.

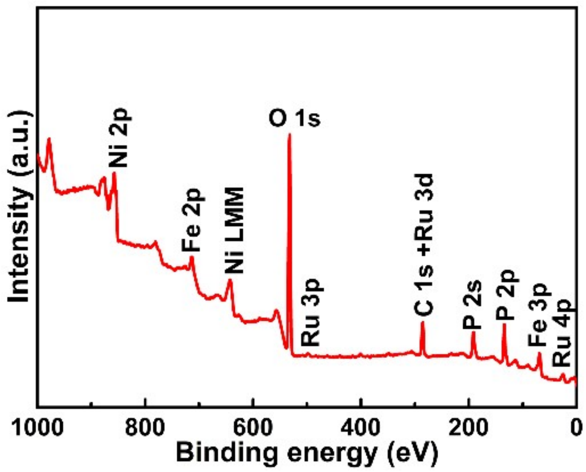

Fig. S10 The survey XPS spectrum for Ni_2P-Fe_2P-Ru_2P/NF.

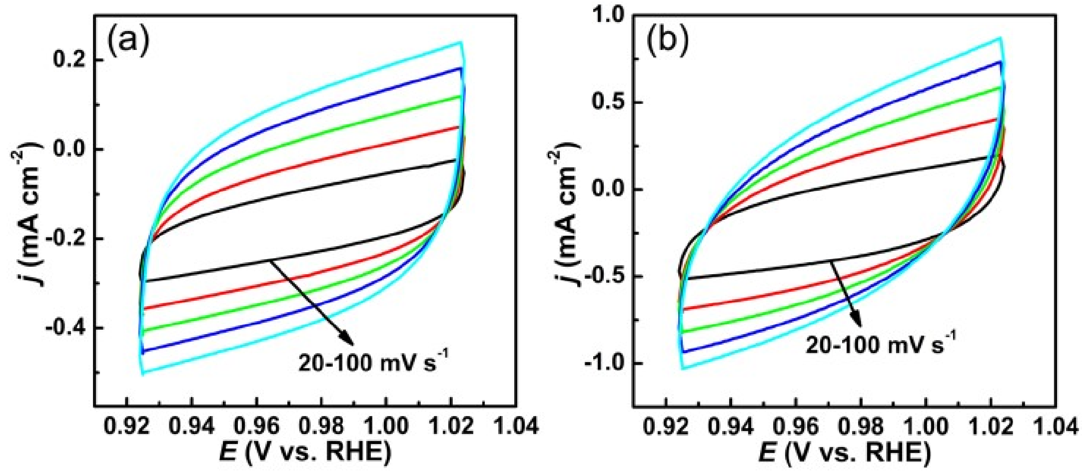

Fig. S11 CV curves of Ni_2P-Fe_2P/NF (a) and Ni_2P-Fe_2P-Ru_2P/NF (b) in the non-Faradaic potential region recorded at different scan rates.

Calculation of ECSA:

$$\mathrm{ECSA} = \frac{C_{dl}}{C_s}$$

$$\mathrm{ECSA}_{\mathrm{Ni_2P\text{-}Fe_2P/NF}} = \frac{2.33\ \mathrm{mF\,cm^{-2}}}{40\ \mu\mathrm{F\,cm^{-2}}} = 58.3\ \mathrm{cm^{-2}}_{\mathrm{ECSA}}$$

$$\mathrm{ECSA}_{\mathrm{Ni_2P-Fe_2P-Ru_2P/NF}} = \frac{4.45\,\mathrm{mF\,cm^{-2}}}{40\ \mu\mathrm{F\,cm^{-2}}} = 111.2\,\mathrm{cm^{-2}}_{\mathrm{ECSA}}$$

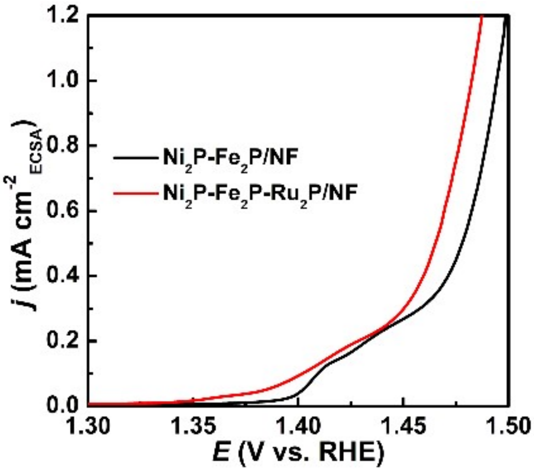

Fig. S12 OER activity of different catalysts in 1 M KOH normalized by ECSA.

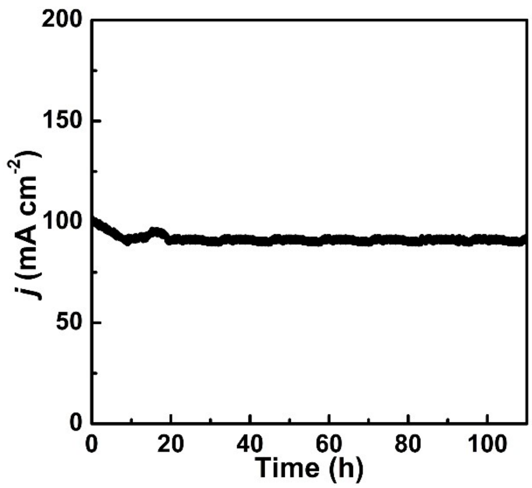

Fig. S13 Durability test of Ni_2P-Fe_2P-Ru_2P/NF at 100 mA cm^{-2} for OER.

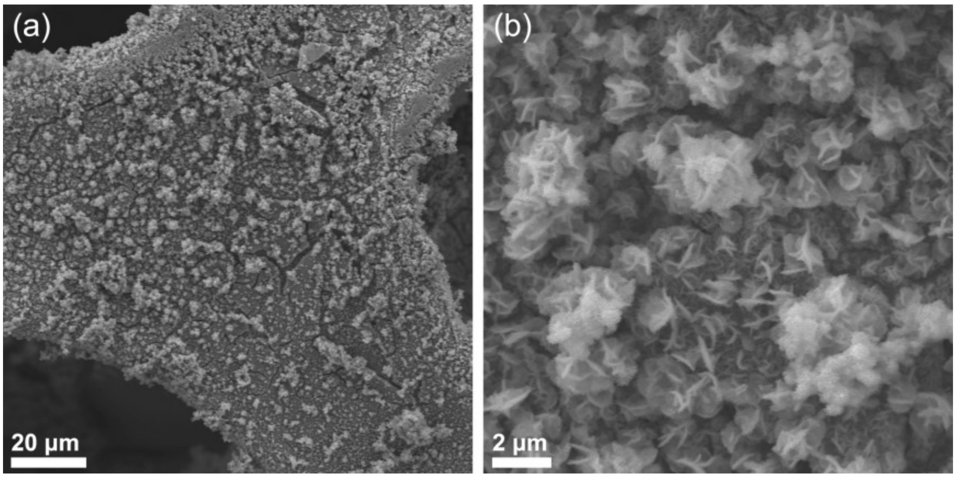

Fig. S14 (a, b) SEM images of Ni_2P-Fe_2P-Ru_2P/NF after OER stability.

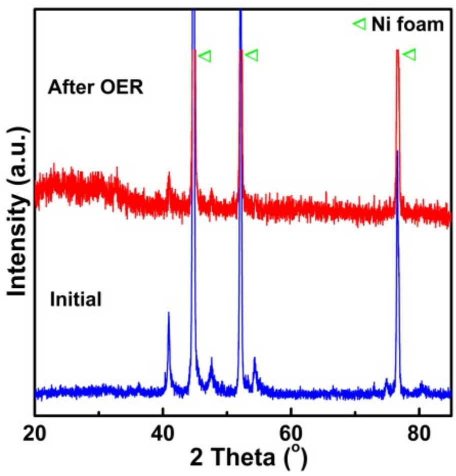

Fig. S15 PXRD patterns of Ni_2P-Fe_2P-Ru_2P/NF before and after stability test for the OER.

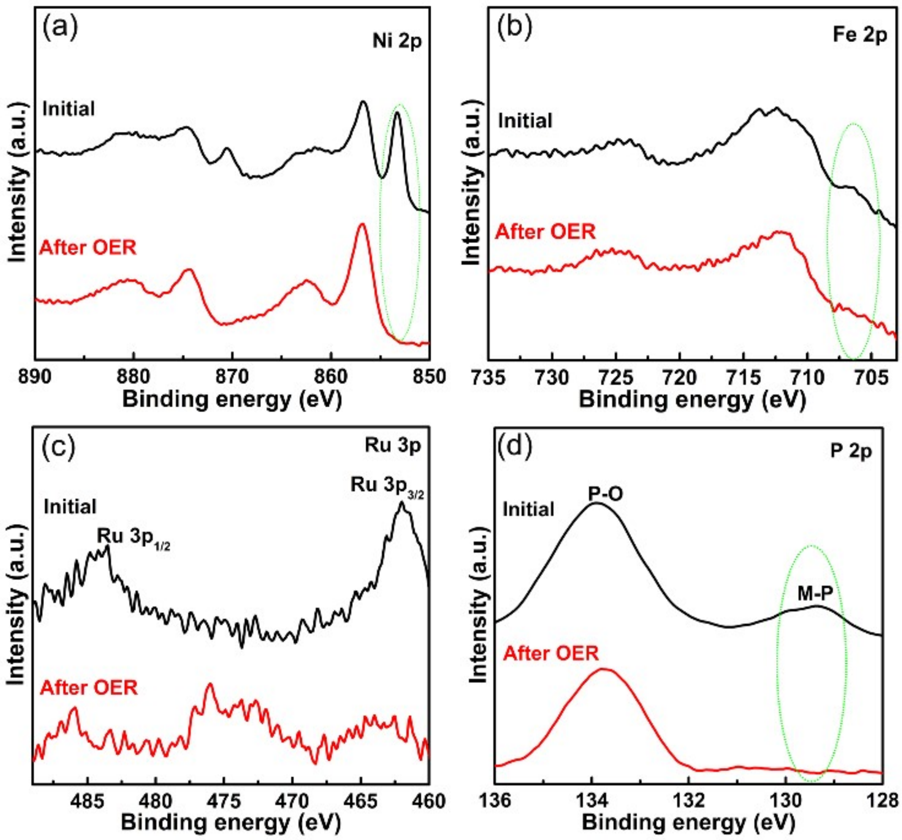

Fig. S16 High-resolution XPS spectra of (a) Ni 2p, (b) Fe 2p, (c) Ru 3p, and P 2p of Ni_2P-Fe_2P-Ru_2P/NF intial and after OER stability testing for the OER.

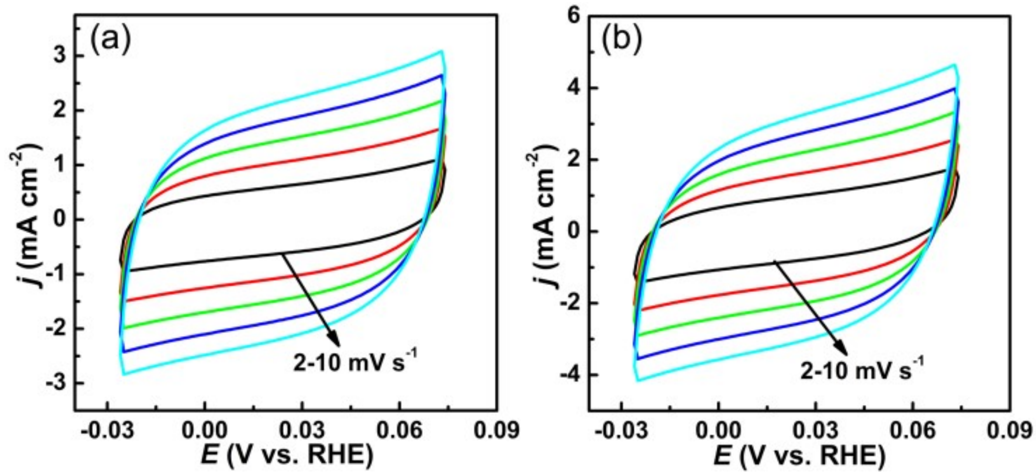

Fig. S17 CV curves of Ni_2P-Fe_2P/NF (a) and Ni_2P-Fe_2P-Ru_2P/NF (b) in the non-Faradaic potential region recorded at different scan rates.

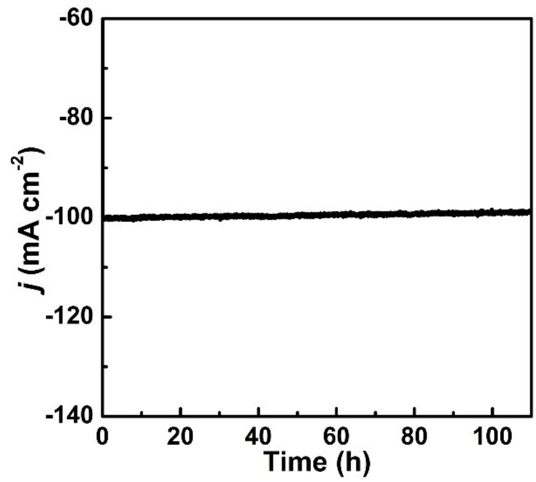

Fig. S18 Durability test of Ni_2P-Fe_2P-Ru_2P/NF at 100 mA cm^{-2} for OER.

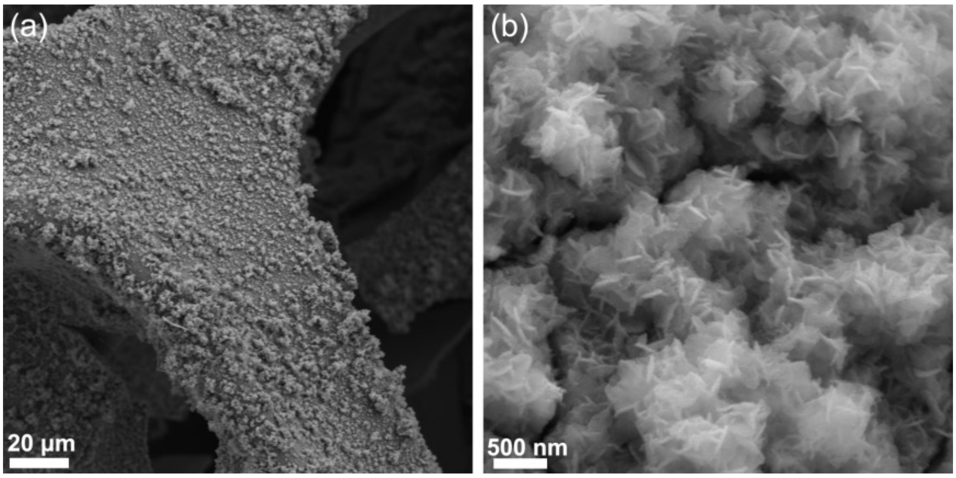

Fig. S19 (a, b) SEM images of Ni_2P-Fe_2P-Ru_2P/NF after the HER stability.

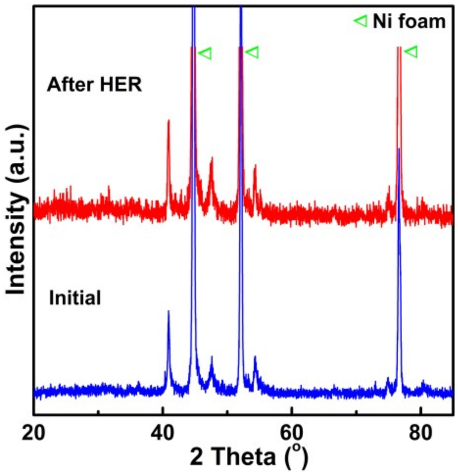

Fig. S20 PXRD patterns of Ni_2P-Fe_2P-Ru_2P/NF before and after stability test for the HER.

Table S1. Comparing the electrocatalytic OER performance of Ni_2P-Fe_2P-Ru_2P/NF with many catalysts recently reported.

Catalysts	Tafel slop (mV dec^{-1})	η_{10} (mV)	Reference
Ni_2P-Fe_2P-Ru_2P/NF	**30.5**	**195**	**This work**
Mo-Ni_3S_2/Ni_xP_y/NF	60.6	/	*Adv. Energy. Mater.* 2020, 10, 1903891.
$NiMoO_x$/NiMoS	34	186	*Nat. Commun.* 2020, 11, 5462.
MoS_2/Co_9S_8/Ni_3S_2/Ni	58	166	*J. Am. Chem. Soc.* 2019, 141, 10417
$CoMoS_4$/Ni_3S_2	63	200	*J. Power Sources* 2019, 416, 95.
Porous Ni_3S_4	67	257	*Adv. Funct. Mater.* 2019, 29, 1900315.
Co_2P NCs	60	280	*Adv. Mater.* 2018, 30, 1705796.
Ni_2P-CoP	69	320	*ACS Appl. Mater. Interfaces* 2017, 9, 23222.
NiCoP/C	96	330	*Angew. Chem. Int. Ed.* 2017, 56, 3897.
MAF-X27-OH	88	292	*J. Am. Chem. Soc.* 2016, 138, 8336

Table S2. Comparing the electrocatalytic HER performance of Ni_2P-Fe_2P-Ru_2P/NF with many catalysts recently reported.

Catalysts	Tafel slop (mV dec^{-1})	η_{10} (mV)	Reference
Ni_2P-Fe_2P-Ru_2P/NF	**85.1**	**78.6**	**This work**
Mo-Ni_3S_2/Ni_xP_y/NF	68.4	109	*Adv. Energy. Mater.* 2020, 10, 1903891.
$CoMoS_4$/Ni_3S_2	169	158	*J. Power Sources* 2019, 416, 95.
Ni_2P-Fe_2P/NF	86	128	*Adv. Funct. Mater.* 2020, 30, 2006484.
NiFeP/NCH	125	216	*J. Am. Chem. Soc.* 2019, 141, 7906.
MoS_2/Co_9S_8/Ni_3S_2/Ni	85	113	*J. Am. Chem. Soc.* 2019, 141, 10417
$(Co_{1-x}Ni_x)$ $(S_{1-y}Py)_2$/G	85	117	*Adv. Energy Mater.* 2018, 8, 1802319.
$Ni_{12}P_5$/$Ni_3(PO_4)_2$-HS	93.1	114	*Appl. Catal. B Environ* 2017, 204, 486.

Table S3. Comparing the electrocatalytic performance of Ni_2P-Fe_2P-Ru_2P/NF with recently reported catalysts for overall water splitting.

Catalysts	Cell voltage (V)	Reference
Ni_2P-Fe_2P-Ru_2P/NF	**1.49**	**This work**
Fe-CoP/$Ni(OH)_2$	1.52	*Adv. Funct. Mater.* 2021, 31, 2101578.
Mo-Ni_3S_2/NixPy/NF	1.46	*Adv. Energy. Mater.* 2020, 10, 1903891.
NiCoP@NiMn LDH/NF	1.51	*Appl. Mater. Interfaces* 2020, 12, 4385.
Cr-doped FeNiP/NCN	1.5	*Adv. Mater.* 2019, 31, 1900178.
NiFeP/SG	1.54	*Nano Energy* 2019, 58, 870.
NiFeP/NCH	1.59	*J. Am. Chem. Soc.* 2019, 141, 7906.
MoS_2/Co_9S_8/Ni_3S_2/Ni	1.54	*J. Am. Chem. Soc.* 2019, 141, 10417
Mo-doped CoP/CC	1.56	*Nano Energy* 2018, 48, 73.