Electronic Supplementary Material (ESI) for Sustainable Energy & Fuels.

Supporting Information

Hierarchical Self-assembly of NiFe-LDH Nanosheets on $CoFe_2O_4$@Co_3S_4 Nanowires for Enhanced Overall Water Splitting

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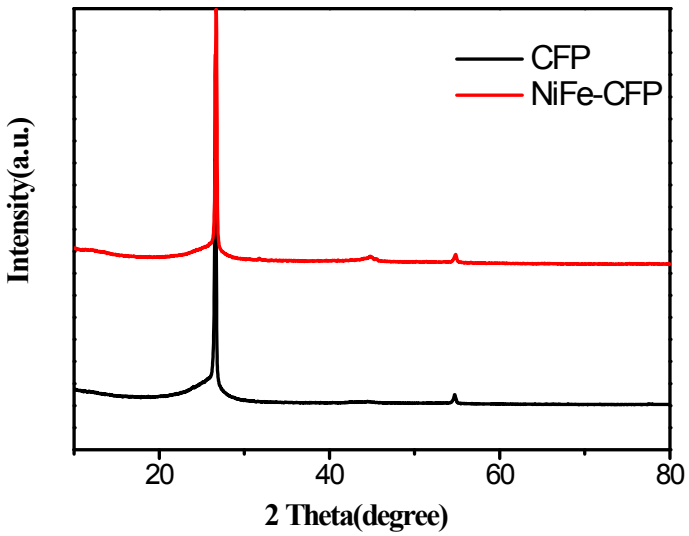

Fig. S1 XRD patterns of the bare carbon fiber paper and NiFe-CFP.

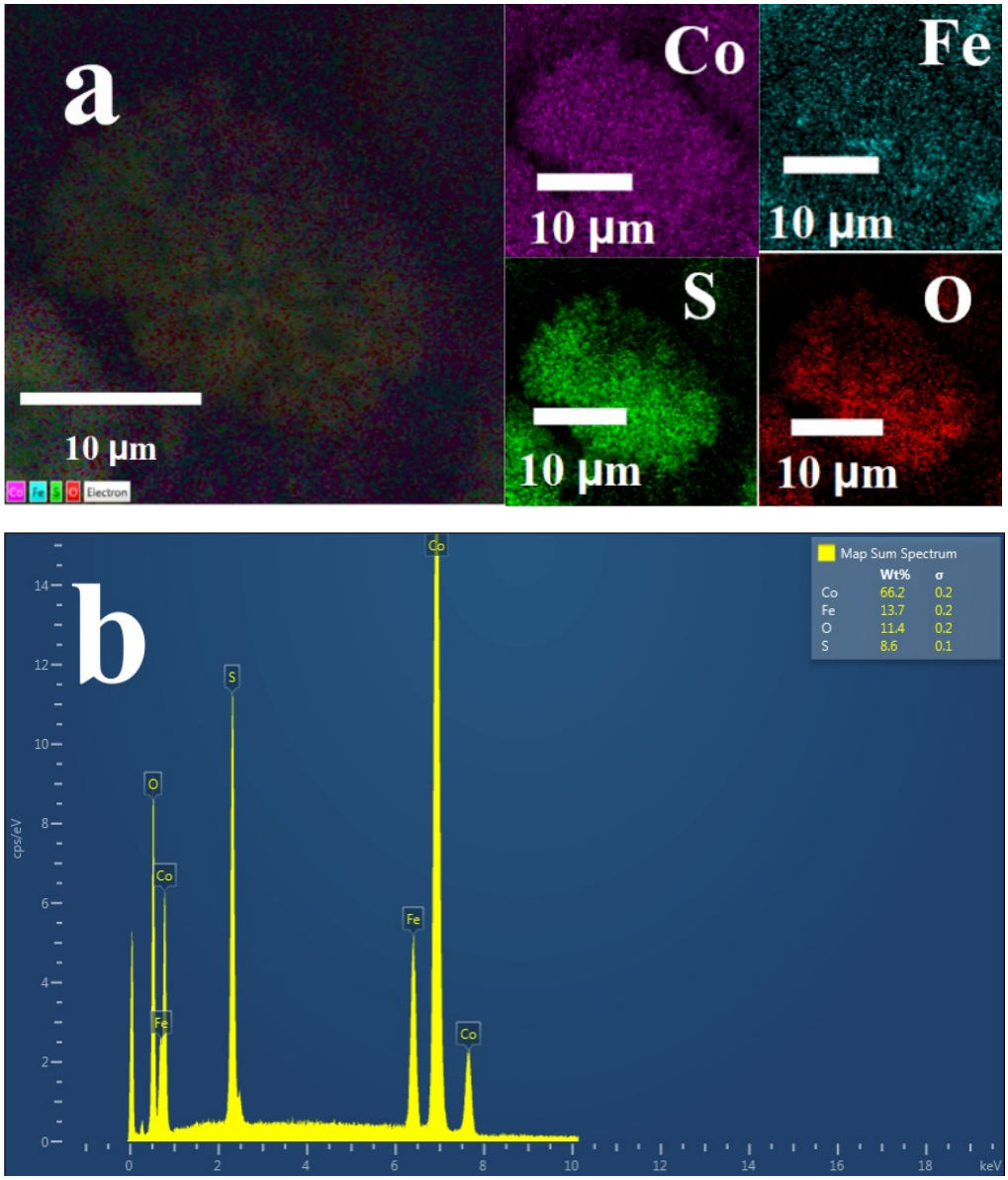

Fig. S2 (a) SEM elemental mapping analysis of $CoFe_2O_4$@Co_3S_4/CFP (b) The corresponding EDX spectrum.

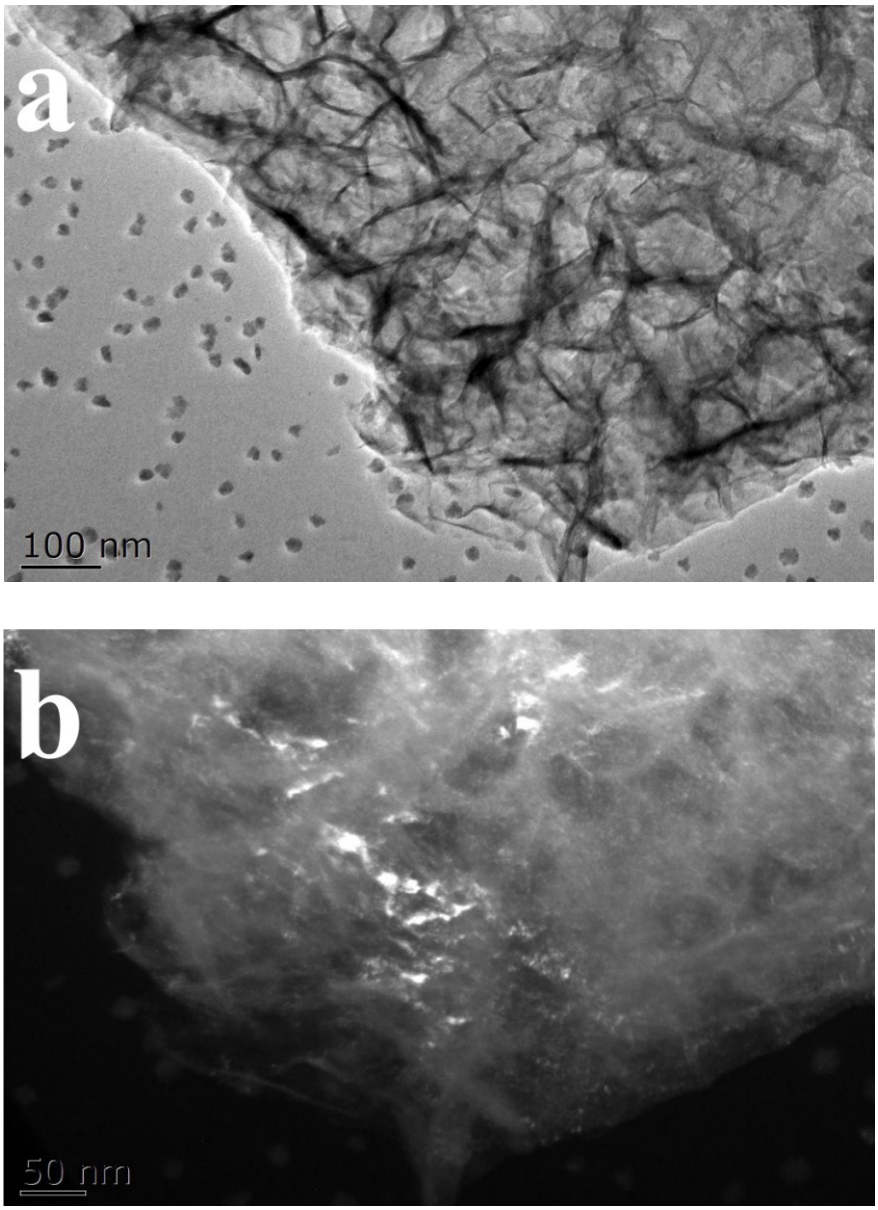

Fig. S3 (a) TEM and (a) high-angle annular dark field scanning transmission electron microscopy (HAADF-STEM) images of NiFe-LDH.

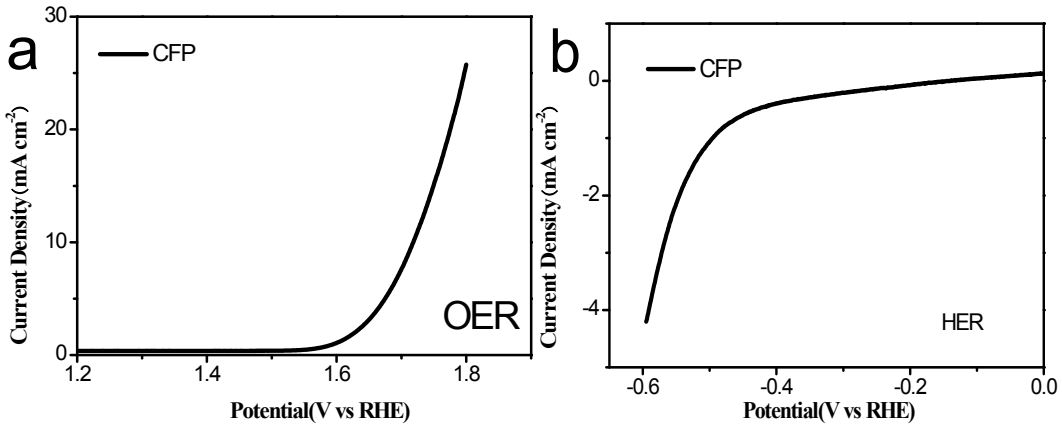

Fig. S4 LSV curves of bare CFP in 1 M KOH to show the background activity for catalyzing OER and HER.

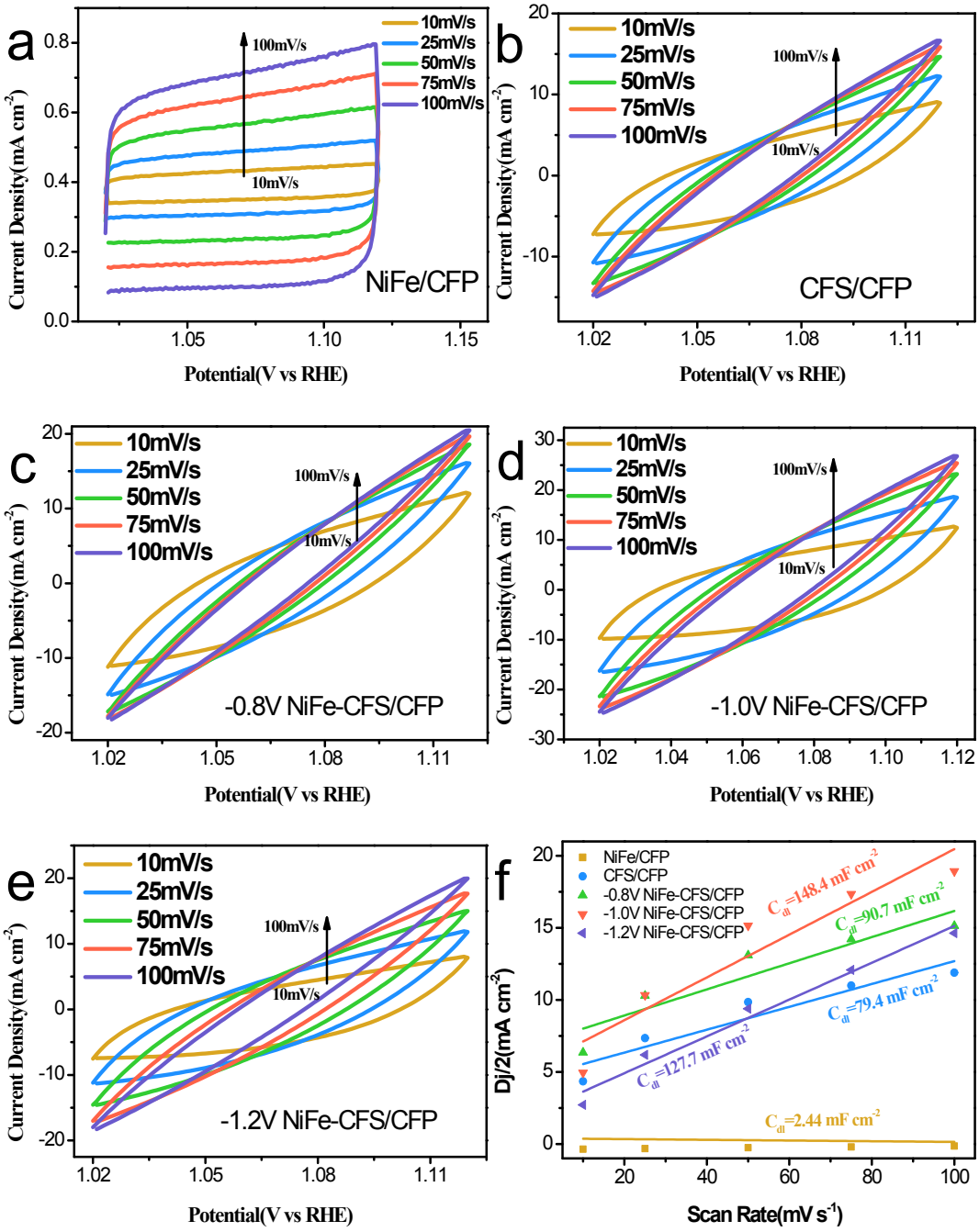

Fig. S5 Cyclic voltammograms of (a) NiFe/CFP, (b) CFS/CFP, (c) -0.8V NiFe-CFS/CFP, (d) -1.0V NiFe-CFS/CFP, and (e) -1.2V NiFe-CFS/CFP composite electrodes between the potential regions of 0.02 and 0.12 V (vs Ag/AgCl) with scan rates of 10, 25, 50, 75, and 100 mV/s toward OER 1 M KOH solution. (f) Current density differences plotted against scan rates under a non-Faradaic range.

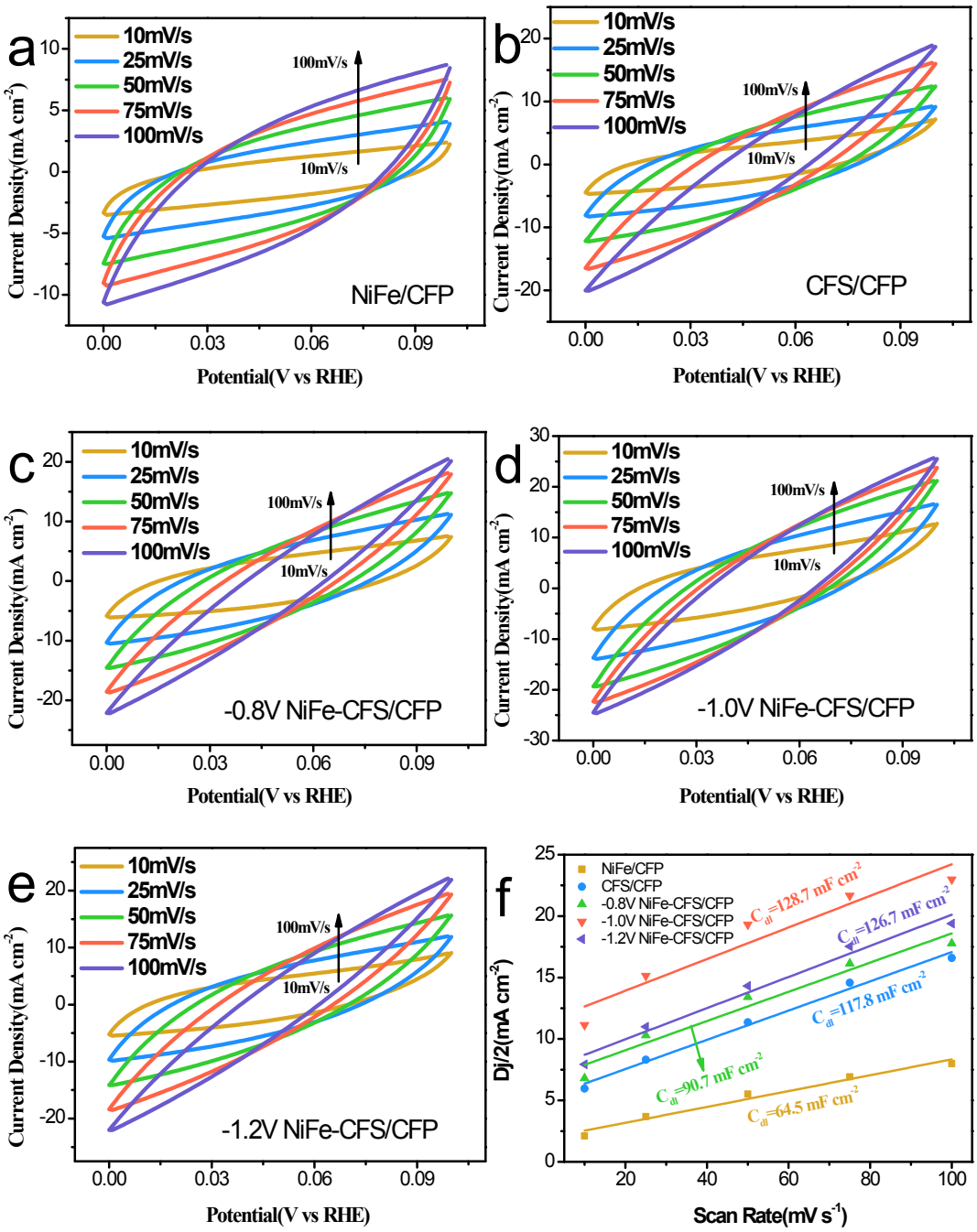

Fig. S6 Cyclic voltammograms of (a) NiFe/CFP, (b) CFS/CFP, (c) -0.8V NiFe-CFS/CFP, (d) -1.0V NiFe-CFS/CFP, and (e) -1.2V NiFe-CFS/CFP composite electrodes between the potential regions of 0 and 0.1 V (vs Ag/AgCl) with scan rates of 10, 25, 50, 75, and 100 mV/s toward HER 1 M KOH solution. (f) Current density differences plotted against scan rates under a non-Faradaic range.

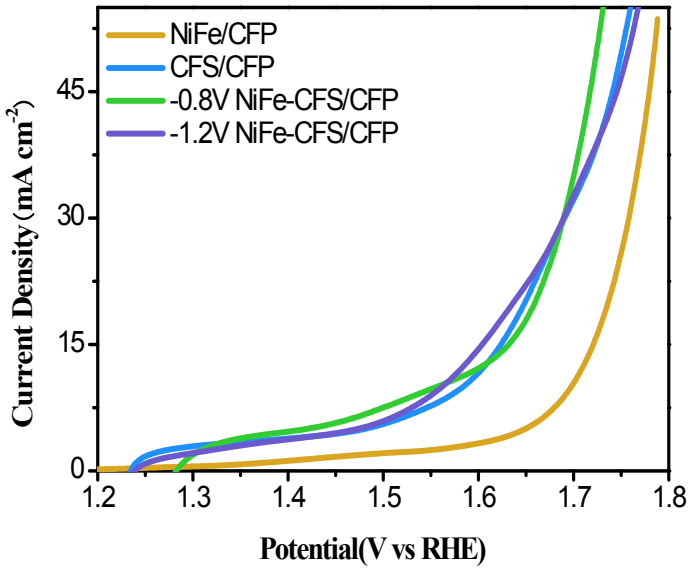

Fig. S7 (a) The polarization curves of NiFe/CFP, CFS/CFP, -0.8V NiFe-CFS/CFP, and -1.2V NiFe-CFS/CFP catalysts in a two-electrode configuration.

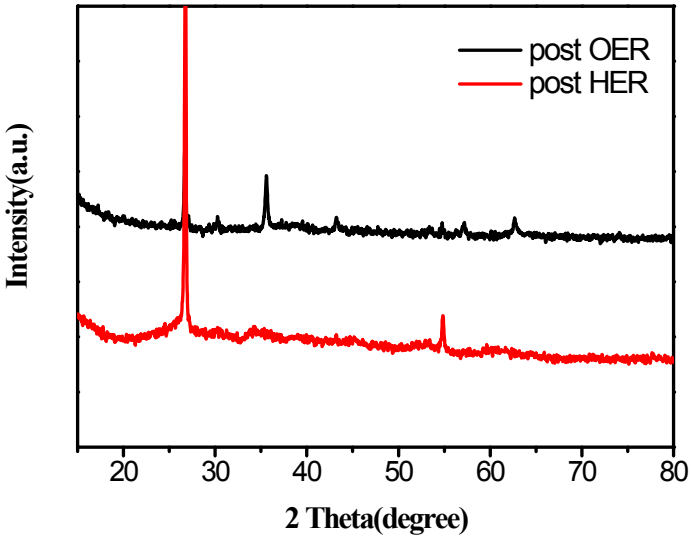

Fig. S8 XRD patterns of post-OER and post-HER of -1.0V NiFe-CFS/CFP electrocatalysts.

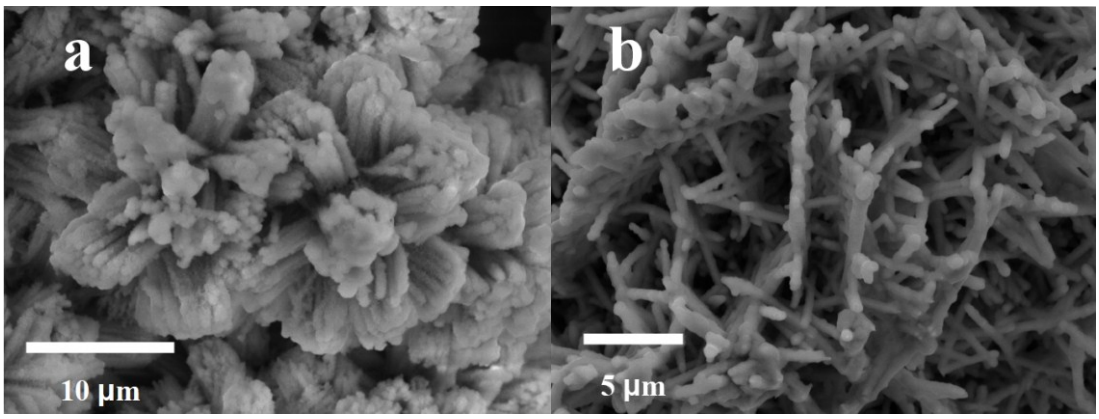

Fig. S9 Low magnification SEM images of -1.0V NiFe-CFS/CFP after long-term (a) OER and (b) HER electrolysis.

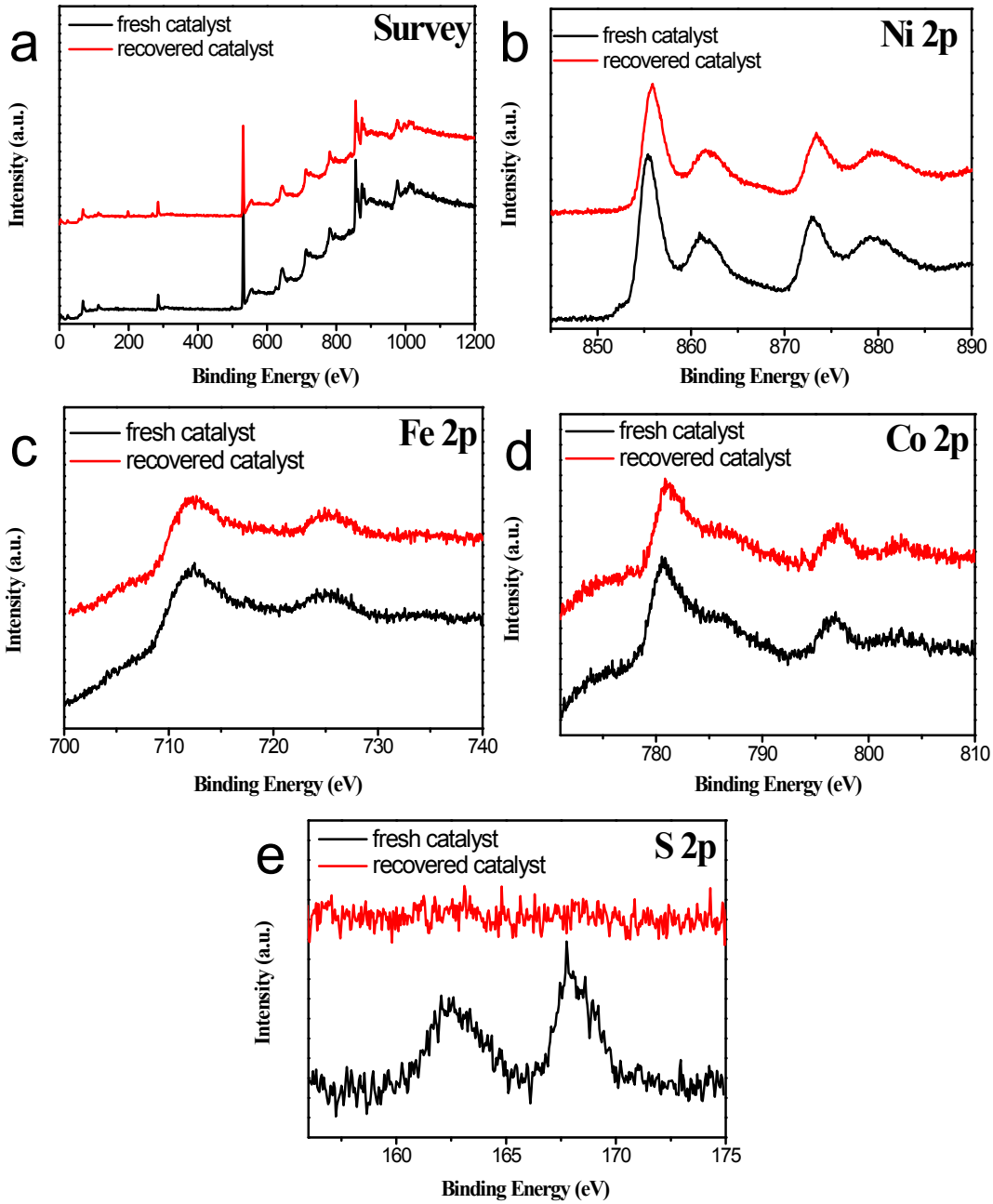

Fig. S10 XPS high-resolution spectra of (a) survey (b) Ni 2p (c) Fe 2p (d) Co 2p and (e) S 2p of fresh and recovered -1.0V NiFe-$CoFe_2O_4$@Co_3S_4/CFP electrocatalysts.

Table S1 Comparison of OER properties for electrocatalysts of catalytic materials

Electrode	Electrolyte	η (mV)	Reference
-1.0V NiFe-CFS/CFP	1 M KOH	233.3 mV@ 50 $mAcm^{-2}$	This work
NiFe/CFP	1 M KOH	292.9 mV@ 50 $mAcm^{-2}$	This work
CFS/CFP	1 M KOH	285.2 mV@ 50 $mAcm^{-2}$	This work
Ru_2O/CFP	1 M KOH	304.8 mV@ 50 $mAcm^{-2}$	This work
Ir_2O/CFP	1 M KOH	445.3 mV@50 mA cm^{-2}	This work
3D porous $CoFe_2O_4$/C	1 M KOH	240 mV@10 mA cm^{-2}	*Adv. Mater.* **2017**, 29, 1604437.
$Co_1Mn_1CH^*$	1 M KOH	322 mV@50 mA cm^{-2}	*J. Am. Chem. Soc.* **2017**, 139, 8320-8328.
$CoFeO_x$/NF	1 M KOH	270 mV@10 mA cm^{-2}	*J. Am. Chem. Soc.* **201**6, 138, 8946-8957.
NiFe/$NiCo_2O_4$/NF	1 M KOH	320 mV@600 mA cm^{-2}	*Adv. Funct. Mater.* **2016**, 26, 3515-3523.
$Ni_3Fe(OH)_9$/NF	1 M KOH	370 mV@100 mA cm^{-2}	*Nat. Commun.* **2015**, 6, 6616.
Carbon-Confined NiCo@$NiCoO_2$	1 M KOH	330 mV@ 10 $mAcm^{-2}$	*Adv. Mater.* **2018**, 1705442
NiFe/Cu_2O NWs/CF	1 M KOH	215 mV@ 10 $mAcm^{-2}$	*ChemSusChem* **2017**, 10, 1475-1481.
$Ni_{0.6}Co_{1.4}P$	1 M KOH	300 mV@ 10 $mAcm^{-2}$	*Adv. Funct. Mater.* **2018**, 28, 1706008
Co-NC@Mo_2C	1 M KOH	347 mV@ 10 $mAcm^{-2}$	*Nano Energy* **2019**, 57, 746
$NiCo_2O_4$/NF*	1 M KOH	420 mV@ 50 $mAcm^{-2}$	*Adv. Funct. Mater.* **2016**, 26, 4661.
NiFe-LDH/CNT	1 M KOH	247 mV@ 10 $mAcm^{-2}$	*J Am Chem Soc.* **2013**, 135, 8452-8455.

Table S2 Comparison of HER properties for electrocatalysts of catalytic materials

Electrode	Electrolyte	η (mV)	Reference
-1.0V NiFe-CFS/CFP	1 M KOH	98 mV@ 10 $mAcm^{-2}$	This work
Pt-C/CFP	1 M KOH	35.8 mV@ 10 $mAcm^{-2}$	This work
$Ni_{0.75}Fe_{0.125}V_{0.125}$-LDHs	1 M KOH	125 mV@ 10 $mAcm^{-2}$	*Small*, **2018**, 14, 1703257.
Ni_5P_4/NiP_2/NiFe LDH	1 M KOH	124 mV@ 10 $mAcm^{-2}$	*J. Mater. Chem. A*, **2018**, 6, 13619-13623.
Co_3O_4@MoS_2	1 M KOH	90 mV@ 10 $mAcm^{-2}$	*J. Mater. Chem. A* **2017**, 28, 2067-2072.
MoS_2 nanowall	1 M NaOH	150 mV@ 10 $mAcm^{-2}$	*Nano Res.* **201**7, 10, 1178-1188.
MoS_2/Ni_3S_2/NF	1 M KOH	110 mV@ 10 $mAcm^{-2}$	*Angew. Chem.* **2016**, 55, 6702-6707
NCNT/Ni-$NiFe_2O_4$/Ni foam	1 M KOH	140 mV@ 10 $mAcm^{-2}$	*Catal. Sci. Technol.* **2019**, 9, 1595-1601
$NiCo_2O_4$/NiFe LDH	1 M KOH	192 mV@ 10 $mAcm^{-2}$	*ACS Appl. Mater. Interfaces* **2017**, 9, 1488.
Cu@NC NT/CF	1 M KOH	123 mV@ 10 $mAcm^{-2}$	*ACS Appl. Mater. Interfaces* **2017**, 9, 36857-36864
NiMoN	1 M KOH	109 mV@ 10 $mAcm^{-2}$	*Adv. Energy Mater.* **2016**, 6, 1600221-1600227.
Fe_3C embedded Fe/N doped Carbon fiber	1 M KOH	86 mV@ 10 $mAcm^{-2}$	*J. Mater. Chem. A* **2017**, 5, 7507-7515.
Co_1Mn_1CH/NF	1 M KOH	180 mV@ 10 $mAcm^{-2}$	*J. Am. Chem. Soc.* **2017**, 139, 8320-8328.

Table S3 Comparison of overall water splitting performance for the recently reported catalysts and ours.

Electrode	Electrolyte	η (mV)	Reference
-1.0V NiFe-FCS/CFP	1 M KOH	1.53 V@ 10 $mAcm^{-2}$	This work
NF@Ni/NiP	1 M KOH	1.61V@ 10 $mAcm^{-2}$	*Adv. Funct. Mater.* **2016**, 26, 3314-3323
Cu@NiFe LDH	1 M KOH	1.54V@ 10 $mAcm^{-2}$	*Energy Environ. Sci.* **2017**, 10, 1820-1827
NC@$CuCo_2N_x$/CF	1 M KOH	1.62V@ 10 $mAcm^{-2}$	*Adv. Funct. Mater.* **2017**, 27, 1704169.
NiFe LDH/NF	1 M KOH	1.70 V@ 10 $mAcm^{-2}$	*Science*, **2014**, 345, 1593-1596.
$Ni_{0.69}Co_{0.31}P$	1 M KOH	1.59V@ 10 $mAcm^{-2}$	*Nanoscale* **2016**, 8, 19129.
Co_1Mn_1CH	1 M KOH	1.68 V@ 10 $mAcm^{-2}$	*J. Am. Chem. Soc.* **2017** 139, 8320-8328.
NiS/Ni_2P/CC	1 M KOH	1.62 V@ 10 $mAcm^{-2}$	*J. Mater. Chem. A* **2018**, 6, 8233.
Co3S4@MoS2	1 M KOH	1.58 V@ 10 $mAcm^{-2}$	*Nano Energy* **2018**, 47, 494.
Ni–Co–P hollow nanobricks	1 M KOH	1.62 V@ 10 $mAcm^{-2}$	*Energy & Environmental Science* **2018**, 11, 872.
$NiCo_2S_4$@NiFe LDH	1 M KOH	1.60 V@ 10 $mAcm^{-2}$	*ACS Appl. Mater. Interfaces* **2017**, 9, 15364-15372.