Supporting Information:

Unique hierarchical NiFe-LDH/Ni/$NiCo_2S_4$ heterostructure arrays on nickel foam for the improvement of overall water splitting activity

Xi-Song Gong,[a,1] Xing Liu[a,1] and Jian Zhou[a,*]

[a]*Chongqing Key Laboratory of inorganic functional materials, College of chemistry, Chongqing normal university, Chongqing, 401331, P.R. China*

Corresponding author:

Prof. & Dr. Jian Zhou

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Prof. & Dr. Jian Zhou

College of chemistry,

Chongqing normal university

Chongqing, 401331,
P.R. China
E−mail: Jianzhou888888@163.com
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[1]These authors contributed equally.

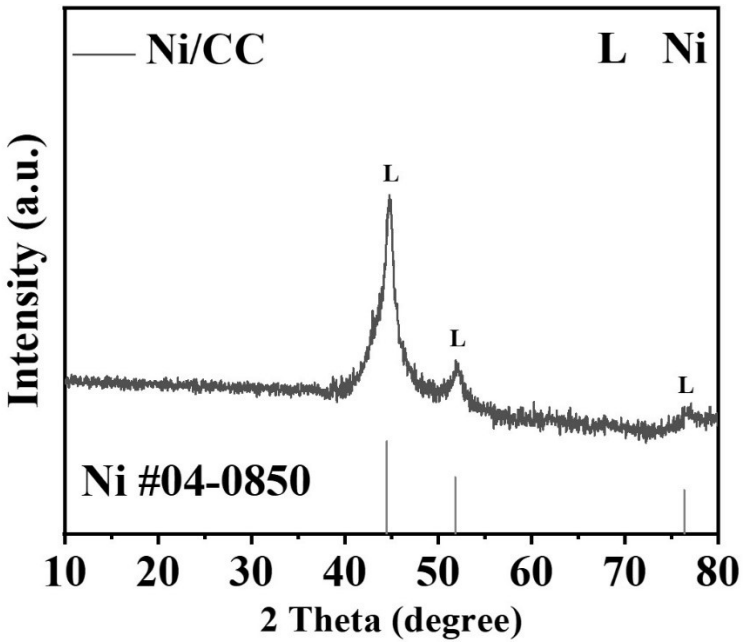

Fig. S1. The XRD patterns of the Ni/CC.

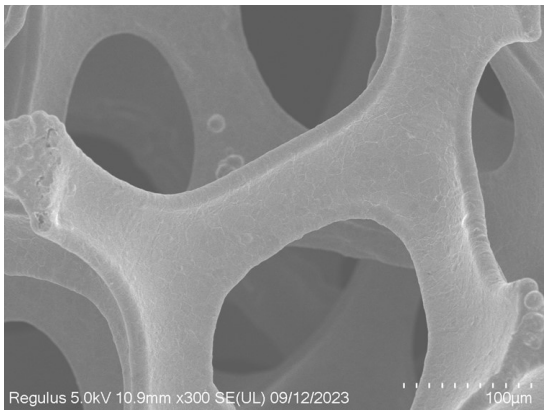

Fig. S2. The SEM of NF.

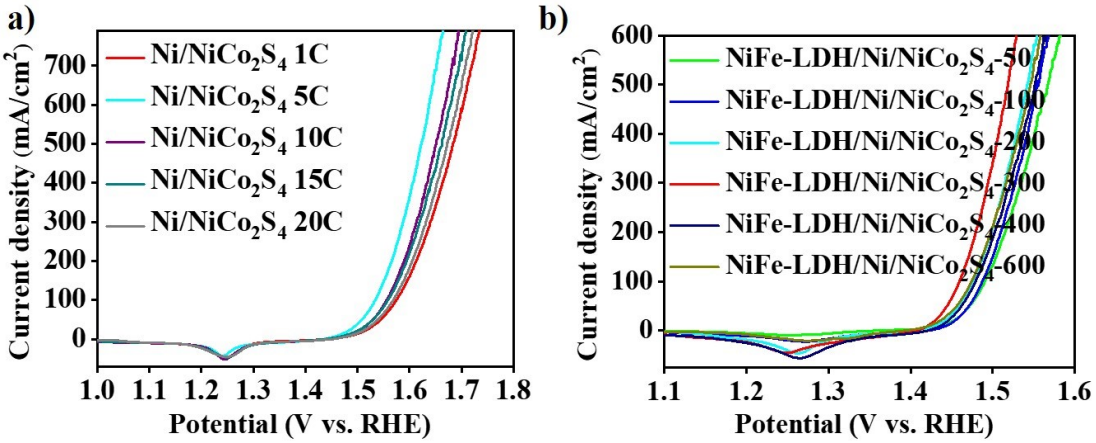

Fig. S3. OER polarization curves of (a) different electrodeposition cycles of the Ni nanoparticles (b) different electrodeposition times of the NiFe-LDH.

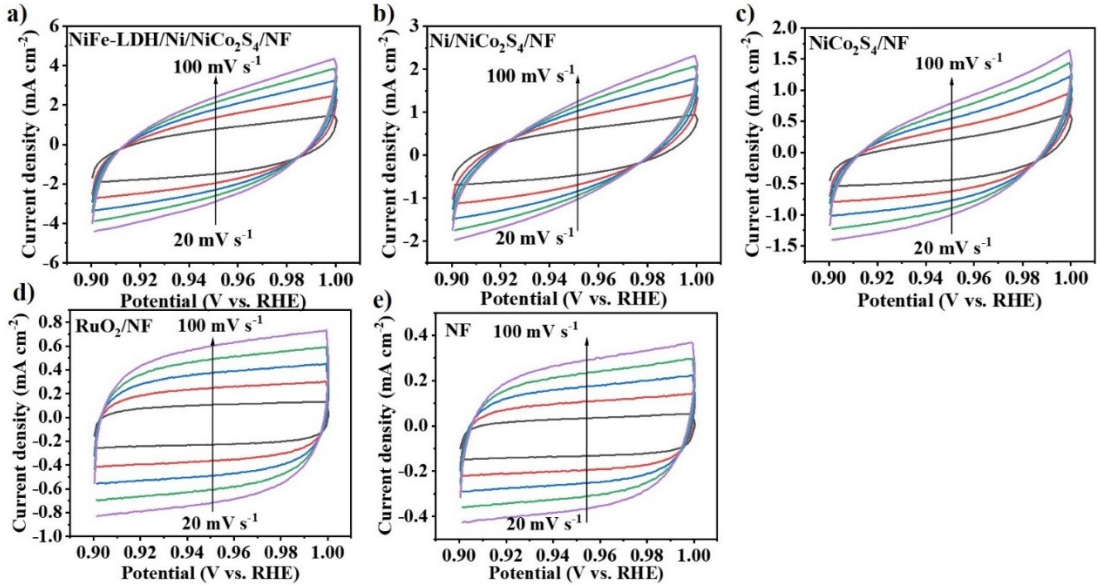

Fig. S4. CV curves for determining electrochemical activity surface area (ECSA) of NiFe-LDH/Ni/$NiCo_2S_4$/NF (a), Ni/$NiCo_2S_4$/NF (b), $NiCo_2S_4$/NF (c), RuO_2/NF (d), and NF (e).

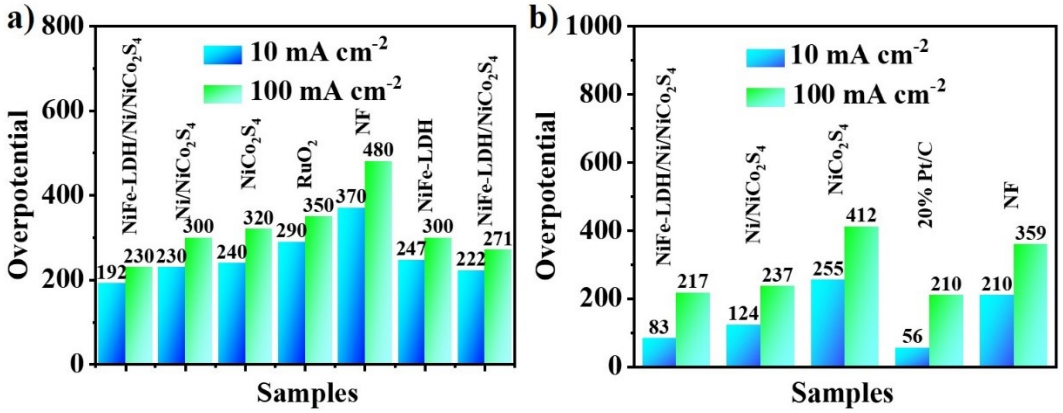

Fig. S5. Overpotential histogram of different samples at different current densities (a) Overpotential for OER, (b) Overpotential for HER.

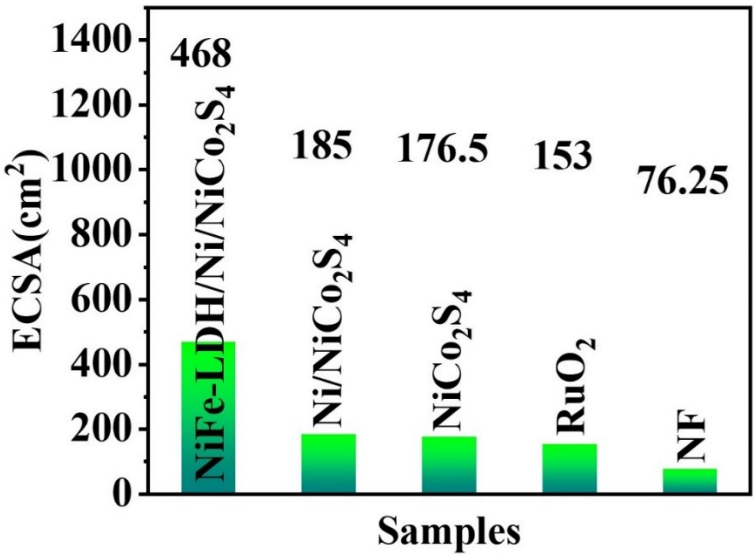

Fig S6. Calculation of ECSA values.

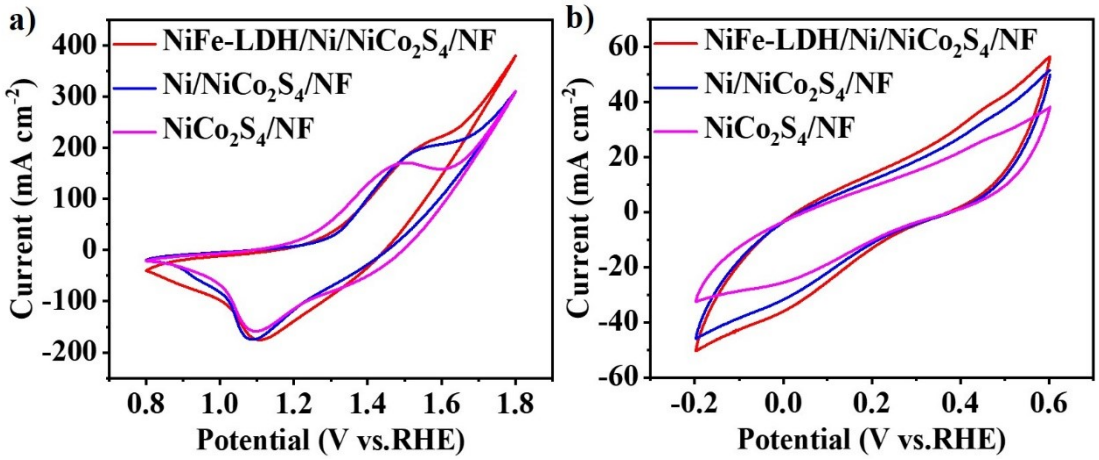

Fig. S7. CV curves of (a) NiFe-LDH/Ni/$NiCo_2S_4$/NF, Ni/$NiCo_2S_4$/NF, and $NiCo_2S_4$/NF in 1.0 M PBS (pH = 6.87) with a scan rate of 50 mV s^{-1}. (b) NiFe-LDH/Ni/$NiCo_2S_4$/NF, Ni/$NiCo_2S_4$/NF, and $NiCo_2S_4$/NF for determining the redox surface sites of Ni^{2+}/Ni^{3+} in 1.0 M KOH with a scan rate of 50 mV s^{-1}.

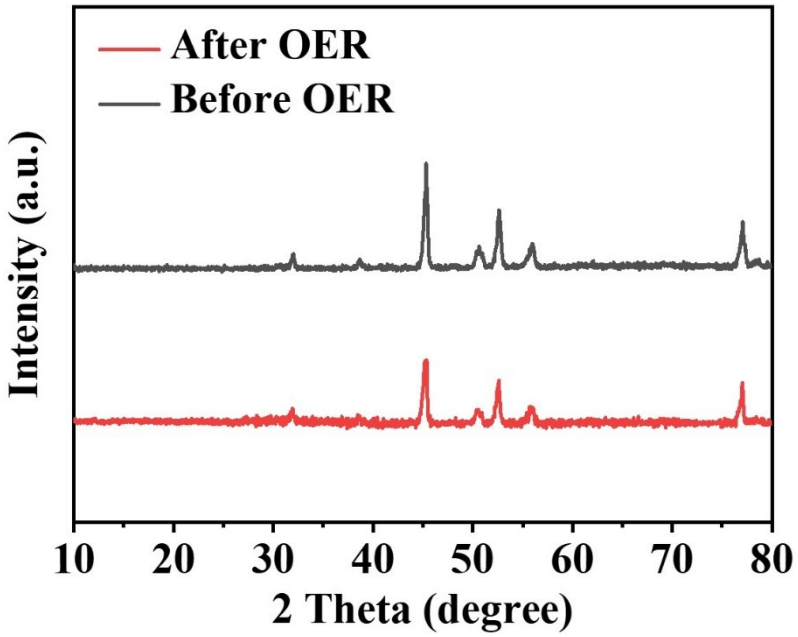

Fig. S8. The XRD of NiFe-LDH/Ni/$NiCo_2S_4$/NF after OER stability test.

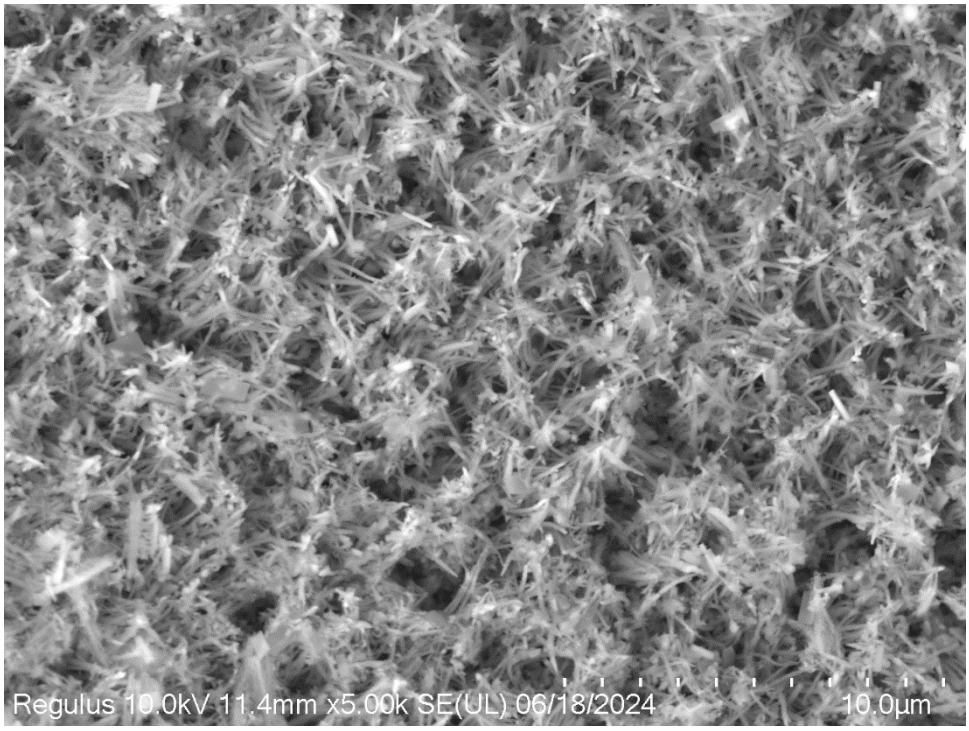

Fig. S9. The SEM image of NiFe-LDH/Ni/$NiCo_2S_4$/NF after OER stability test.

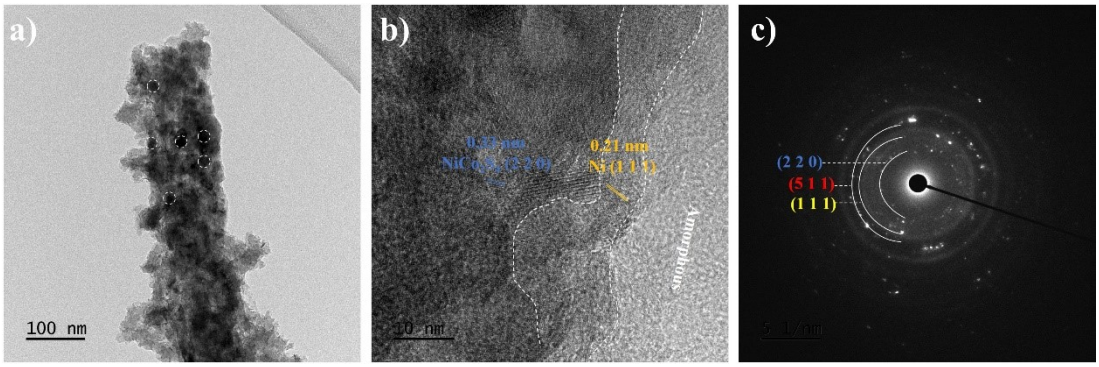

Fig. S10. The TEM and HRTEM images (a-b) of NiFe-LDH/Ni/$NiCo_2S_4$/NF after OER stability test, (c) SAED pattern of NiFe-LDH/Ni/$NiCo_2S_4$.

Table S1 Comparison of OWS performance of the reported electrocatalysts.

Catalysts	Substrate	Electrolyte	Voltage (V)	Current density ($mA \cdot cm^{-2}$)	Ref
NiFe-LDH/Ni/$NiCo_2S_4$	NF	1.0 M KOH	1.53	10	This work
Ni@NiFe LDH	NF	1.0 M KOH	1.53	10	[1]
MoO_3/Ni-NiO	CC	1.0 M KOH	1.55	10	[2]
$NiCo_2S_4$@NiFe LDH	NF	1.0 M KOH	1.60	10	[3]
Ni/MoO_2@CN	NF	1.0 M KOH	1.73	200	[4]
$NiCoSe_2$@NiO@$CoNi_2S_4$@CoS_2	NF	1.0 M KOH	1.583	100	[5]
$Ni_{0.75}Fe_{0.125}V_{0.125}$LDHs	NF	1.0 M KOH	1.591	10	[6]
POM@ZnCoS	NF	1.0 M KOH	1.56	10	[7]
CdFe-BDC	NF	1.0 M KOH	1.68	10	[8]
NiFe LDH@NiCoP	NF	1.0 M KOH	1.57	10	[9]
NiFe-LDH/$NiCo_2O_4$	NF	1.0 M KOH	1.60	10	[10]
FeOOH/$NiCo_2S_4$/Ni_3S_2	NF	1.0 M KOH	1.55	10	[11]
CoP/Co_3O_4	TM	1.0 M KOH	1.59	10	[12]
MnO_x/NiCoP	NF	1.0 M KOH	1.59	10	[13]
N-doped Fe_2O_3/$NiTe_2$	NF	1.0 M KOH	1.54	10	[14]
$Co_{0.15}$@ARC	CC	1.0 M KOH	1.64	10	[15]

Table S2. The resistance value of each component in the equivalent electrical circuit.

Samples	R_s (Ω)	R_{ct} (Ω)
NiFe-LDH/Ni/$NiCo_2S_4$/NF	1.87	0.52
Ni/$NiCo_2S_4$/NF	1.91	3.85
NiFe-LDH/$NiCo_2S_4$/NF	1.95	1.96
$NiCo_2S_4$/NF	2.00	5.22
NF	1.98	45.01
RuO_2	1.63	13.76

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