Supporting Information

Crystalline Multi-Metal Nanosheets Array with Enriched Oxygen Vacancies as Efficient and Stable Bifunctional Electrocatalysts for Water Splitting

Muhammad Imran Abdullah,[c] Asima Hameed,[c] Tuoping Hu,[a] Ning Zhang,*[b] and Mingming Ma*[a,c]

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[a] Department of Chemistry, College of Science, North University of China, Taiyuan 030051, China

[b] Department of Biological and Environmental Engineering, Hefei University, Hefei, Anhui 230022, China

[c] CAS Key Laboratory of Soft Matter Chemistry, Hefei National Laboratory for Physical Sciences at Microscale,

University of Science and Technology of China, Hefei, Anhui 230026, China

E-mail: mma@ustc.edu.cn, zhn@hfuu.edu.cn

Catalog

Figure S1. Brunauer-Emmett-Teller (BET) Analysis.

Figure S2. XPS spectrum for CoFeNiO-NSA sample.

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Table S4. The amount of elements in a CoFeNiO-NSA.

Table S5. The amount of elements in a CoFeNi-NSA.

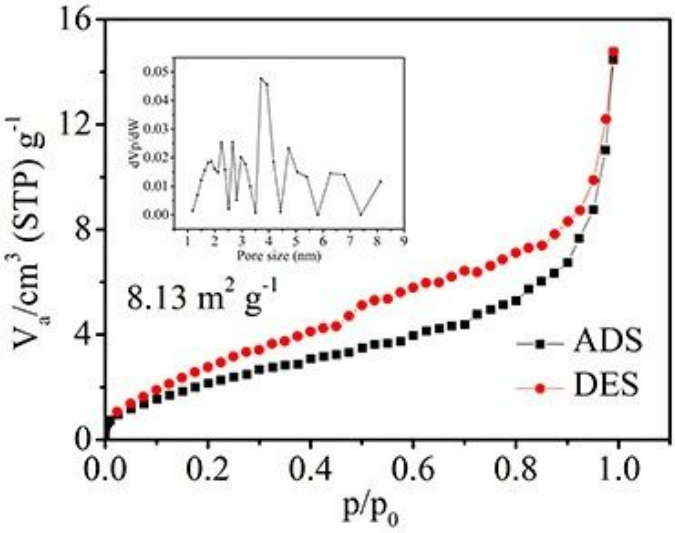

Figure S1. Brunauer-Emmett-Teller (BET) Surface Area Analysis of CoFeNiSe Precursor.

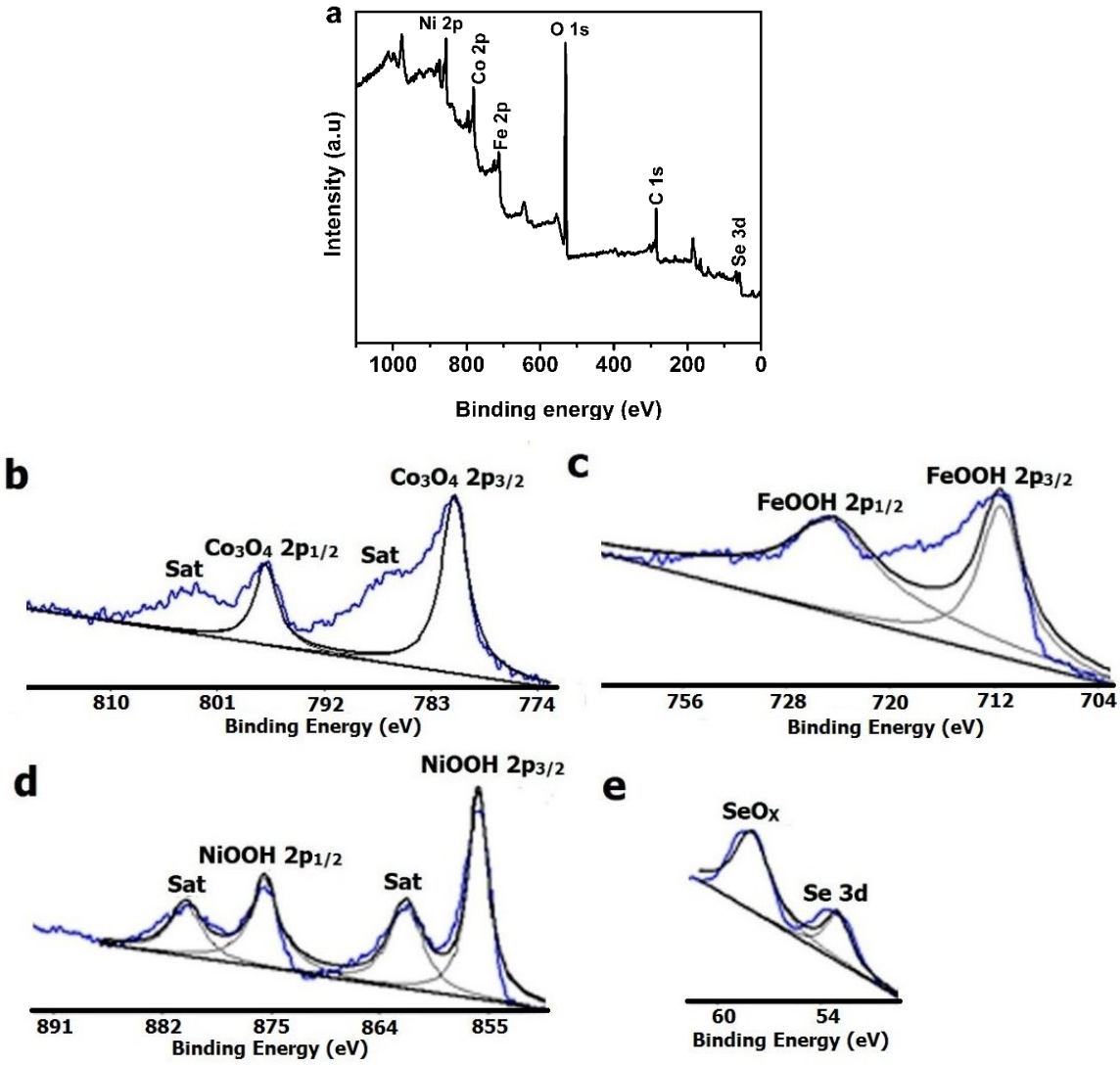

Figure S2. (a)XPS survey spectrum for CoFeNiO-NSA; Core level of XPS spectrum in the (b) Co 2p, (c) Fe 2p, (d) Ni 2p and (e) Se 3d for CoFeNiO-NSA.

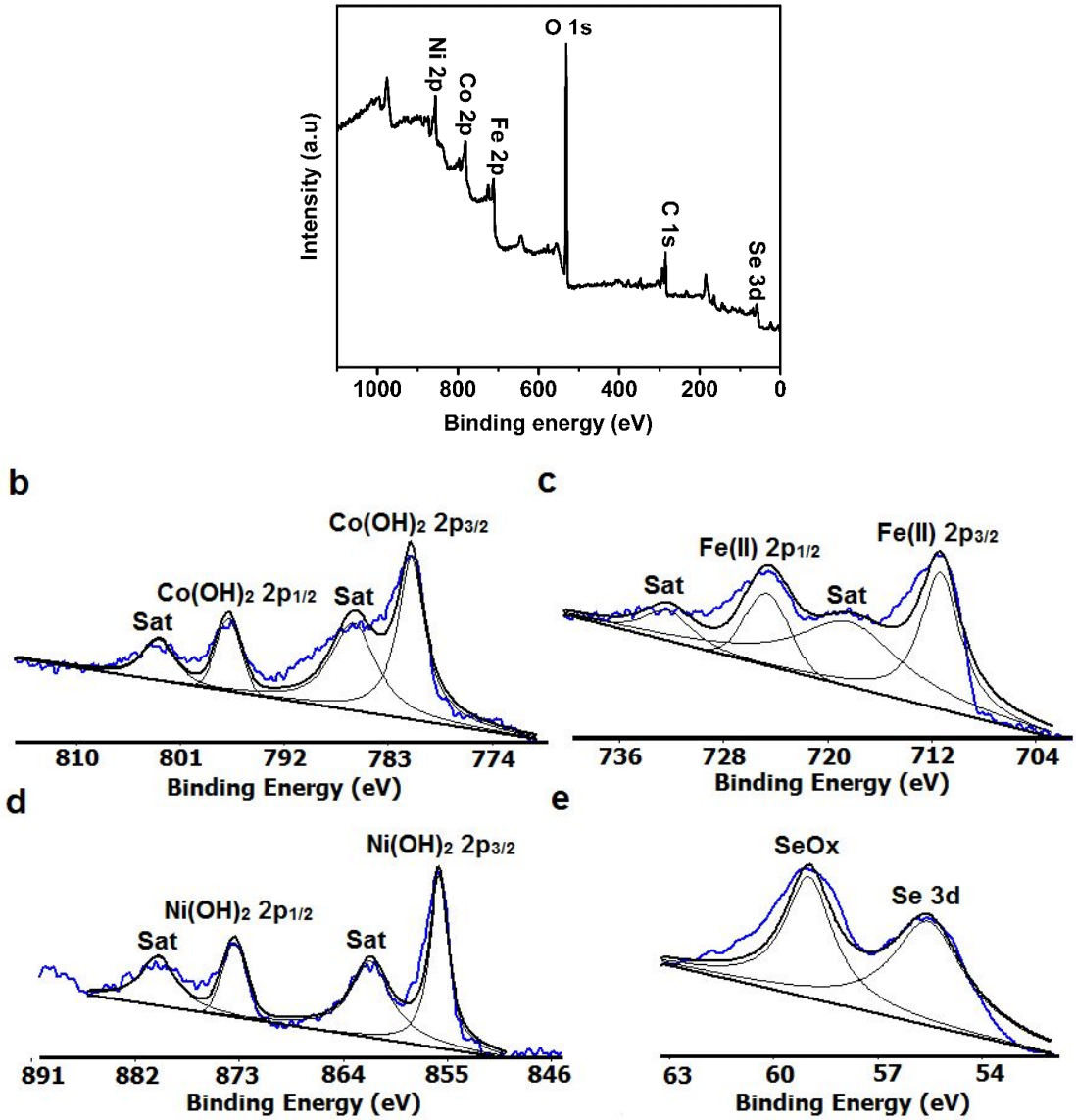

Figure S3 (a)XPS survey spectrum for CoFeNi-NSA; Core level of XPS spectrum in the (b) Co 2p, (c) Fe 2p, (d) Ni 2p and (e) Se 3d for CoFeNi-NSA.·

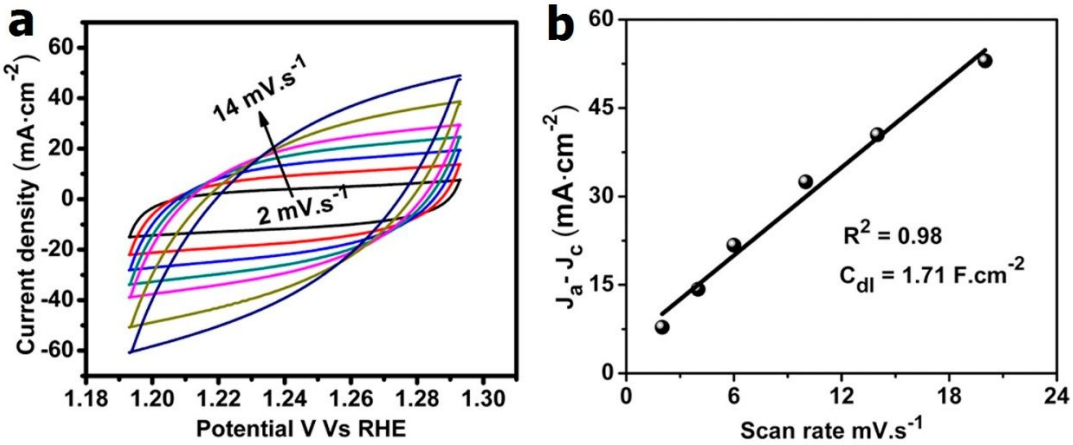

Figure S4 (a) Cyclic voltammograms recorded for the CoFeNiO-NSA electrode at various scan rates in the non-Faradaic region in 1.0 M KOH. (b) Scan rate dependence of the current densities of the CoFeNiO-NSA electrode at 1.24 V vs RHE. The slope of the lines in (b) is the effective electrochemically active surface area of CoFeNiO-NSA electrodes.

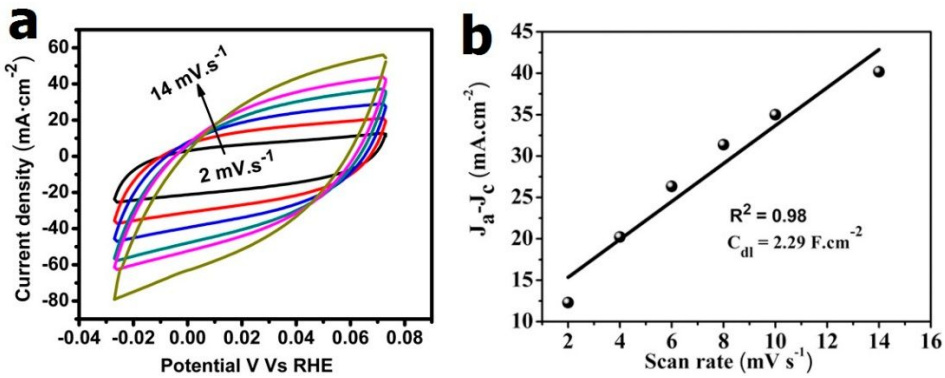

Figure S5 (a) Cyclic voltammograms recorded for the CoFeNi-NSA electrode at various scan rates in the non-Faradaic region in 1.0 M KOH. (b) Scan rate dependence of the current densities of the CoFeNi-NSA electrode at 0.02 V vs RHE. The slope of the lines in (b) is the effective electrochemically active surface area of CoFeNi-NSA electrodes.

Table S1. Comparison of CoNiFe-HNP and other non-noble metal electrocatalysts for HER in 1 M KOH.

Material	Overpotential η(V) @corresponding j	Current density j(mA·cm-2)	Tafel slope (mV –dec-1)	Preparation method	Reference
MoB	0.240	20	59	Commercial	[1]
CoP nanowire	0.209	10	129	Hydrothermal	[2]
Co-NRCNTs	0.37	10	69	Hydrothermal	[3]
MoP nanoparticles	0.13	10	48	Sintering	[4]
NiO/Ni@CNT	0.8	10	82	Anneanling	[5]
MoC	0.151	10	59	Anneanling	[6]
MoxC-Ni@NCV	0.126	10	93	Thermolysis	[7]
CoOx@CN	0.232	10	115	Hydrothermal	[8]
Ni-MoS2	0.98	10	60	Pyrolysis	[9]
Co1Mn1CH/NF	0.180	10	60	Hyrothermal	[10]
NiS-$Ni_2P_2S_6$	0.140	10	72.8	Calcination	[11]
CuNDs/Ni_3S_2 NTs CFs	0.128	10	76.2	Electrodeposition	[12]
Ni_2P NPs/C	0.073	10	73	Photochemical	[13]
CoNiP-NCP	0.058	10	57	Anneanling	[14]
CoFeNi-NSA	**0.049**	**10**	**57**	Eletcrodeposition	**This work**
	0.145	**100**			
	0.175	**200**			

Table S2. Comparison of CoNiFeO-HNP and other non-noble metal electrocatalysts for OER in 1.0 M KOH

Material	Overpotential η(V) @corresponding j	Current density j(mA·cm-2)	Tafel slope (mV –dec-1)	Preparation Methods	Reference
Co_3O_4 NWs	0.42	20	72	Hydrothermal	[15]
γ-CoOOH	0.3	10	38	Hydrothermal	[16]
Co-Fe-P	0.217	10	40	Solvothermal	[17]
Fe0.1Ni0.9O	0.297	10	37	Solvothermal	[18]
NiFe/NF	0.270	10	28	Electrodeposition	[19]
Co_4N	0.257	10	44	Hydrothermal	[20]
Ni3S2/NF	0.260	10	93	Hydrothermal	[21]
CoP nanorods	0.290	10	65	Electrodeposition	[22]
$CoSe_2$ sheets	0.470	10	64	Thermal Process	[23]
Co-Bi NS/G	0.29	10	53	Chemical Reduction	[24]
FeOOH/Co/FeOOH	0.25	20	32	Electrodeposition	[25]
CoMnP	0.33	10	61	Solvothermal	[26]
Co-P film	0.345	10	47	Electrodeposition	[27]
	0.413	100			
	0.463	500			
h-NiSx	0.180	10	96	Electrodeposition Low Temperature sulfurization	[28]
	0.217	100			
	0.316	500			
CoFeBO	0.240	10	53	Electrodeposition	[29]
	0.280	100			
	0.310	500			
G-CoFeW (without iR)	0.191	10	37	Sol-gel hyrolysis	[30]
	0.260	100			
CoNiFeB	0.225	10	40	Chemical redduction phosphorization	[31]
CoFeMo	0.277	10	27.74	solvothermal	[32]
NiS-$Ni_2P_2S_6$	0.220	10	83.2	calcination	[11]
MoFe:NiOOH/$Ni(OH)_2$	0.280	100	47	Hydrothermal	[33]
Fe-Co-P	0.252	10	33	Hydrothermal	[34]

P-Co3O4/NF	0.260	20	60	Annealing	[35]
CS-NiFeCu	0.180	10	33	Electrodeposition	[36]
CoFeNiO-NSA	0.104	10	44	Eletcrodeposition	This Work
	0.240	100			

Table S3. Comparison of CoNiFeO-HNP II CoNiFe-HNP and other non-noble metal electrocatalysts for overall water splitting in 1.0 M KOH.

Material	Voltage (V)	Current density j(mA·cm-2)	Preparation Methods	Reference
NiSe NWs ‖ NiSe NWs	1.63	10	Hydrothermal	37
Ni5P4 ‖ Ni5P4	~1.70	10	Hydrothermal	38
CoP ‖ CoP	1.74	100	Electrodeposition	39
CoP-MNA ‖ CoP-MNA	1.62	10	Electrodeposition	40
CoMnO@CN ‖ CoMnO@CN	1.7	54	Hydrothermal Calcination	41
	1.8	108		
NiFeOx ‖ NiFeOx	1.7	30	Hydrothermal	42
	1.8	100		
EG/$Co_{0.85}$Se/NiFe-LDH ‖ EG/$Co_{0.85}$Se/NiFe-LDH	1.67	10	Hydrothermal	43
	1.71	20		
NiFeS ‖ NiFeS	1.62	10	Hydrothermal	44
CoMnCH ‖ CoMnCH	1.68	10	Hydrothermal	10
AlCoP ‖ AlCoP	1.56	10	Hydrothermal	45
NiS-$Ni_2P_2S_6$ ‖ NiS-$Ni_2P_2S_6$	1.64	10	Calcination	11
CoFeBO NS ‖ Co NS	1.50	10	Eletcrodeposition	29
$P_{8.6}$-Co_3O_4 ‖ $P_{8.6}$-Co_3O_4	1.63	10	Annealing	35
CoP/NCNHP ‖ CoP/NCNHP	1.64	10	Pyrolysis	46
Co_3O_4 UNA ‖ $Co(OH)_2$ UNA	1.69	50	Eletcrodeposition	47
CoFeNiO-NSA ‖ CoFeNi-NSA	1.39	10	Eletcrodeposition	This work
	1.61	50		
	1.69	100		

Table S4. The amount of elements in a CoFeNiO-NSA (in: %Atomic)

	Co	Ni	Fe	Se	Co:Ni:Fe:Se
CoFeNiO-NSA before OER	0.1072	0.0996	0.0870	0.1418	1.23:1.14:1:1.63
post-OER	0.0203	0.0245	0.0206	0.0089	1:1.19:1:0.43

Table S5. The amount of elements in a CoFeNi-NSA (in: %Atomic)

	Co	Ni	Fe	Se	Co:Ni:Fe:Se
CoFeNi-NSA	0.0810	0.1058	0.0747	0.1401	1.08:1.41:1:1.87
post-HER	0.0645	0.0841	0.0644	0.0081	1:1.30:1:0.12

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