Electronic Supplementary Material (ESI) for Dalton Transactions.

Electronic Supplementary Information

Sulfur Doped FeO_x Nanosheet Arrays Supported on Nickel Foam for Efficient Alkaline Seawater Splitting

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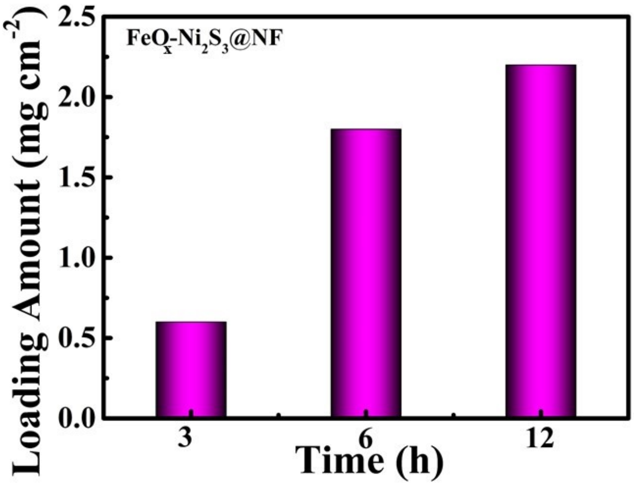


Figure S1. The loading amount of FeO_x-Ni_3S_2 catalyst material on NF at 3 h, 6 h and 12 h, respectively.

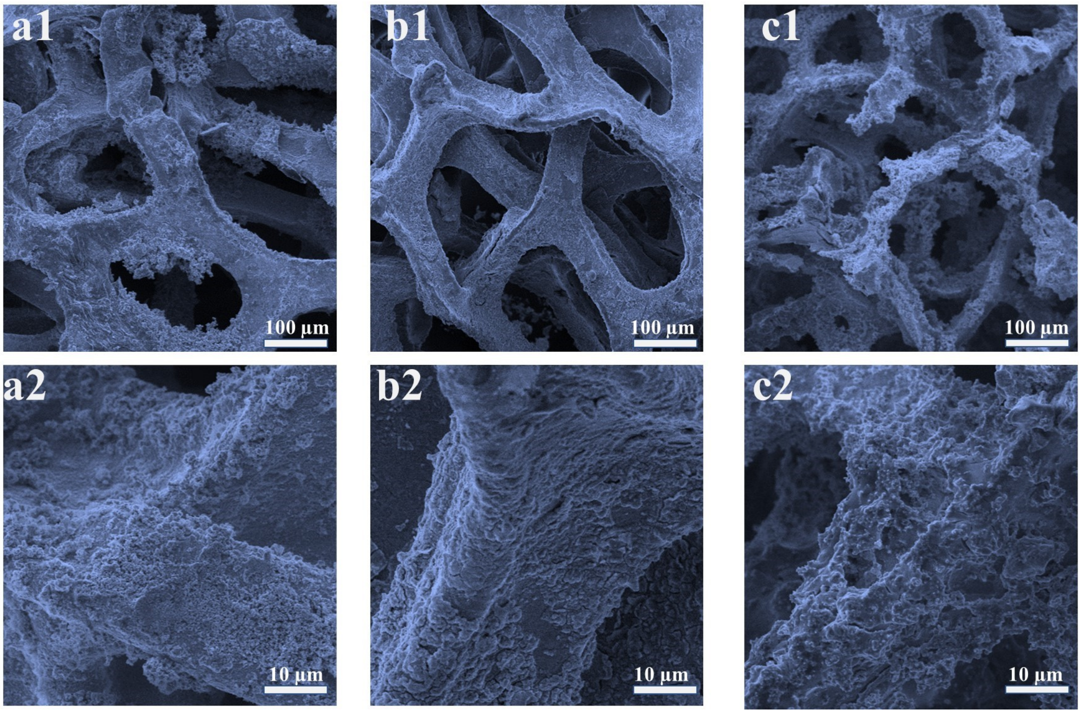


Figure S2. FESEM images of the FeO_x-Ni_3S_2@NF composite with hydrothermal for (a) 3 h, (b) 6 h and (c) 12 h.

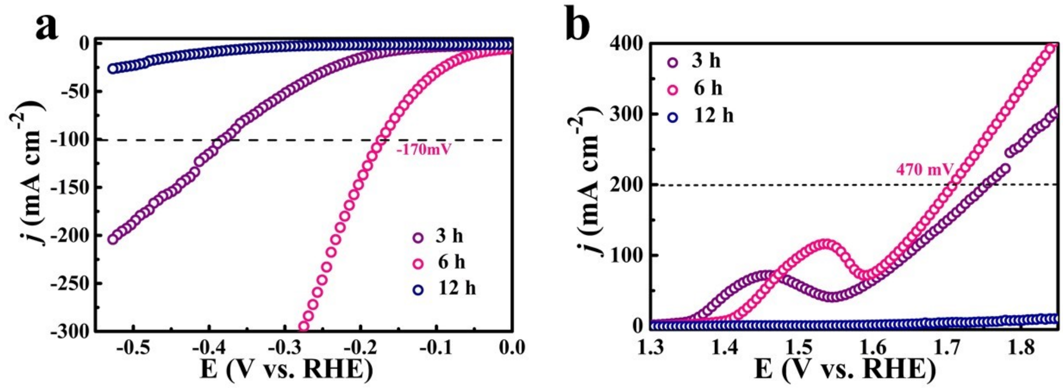


Figure S3. LSV curves of FeO_x-Ni_3S_2@NF for HER (a) and OER (b) in 1.0 M KOH+0.5 M KOH with different hydrothermal time.

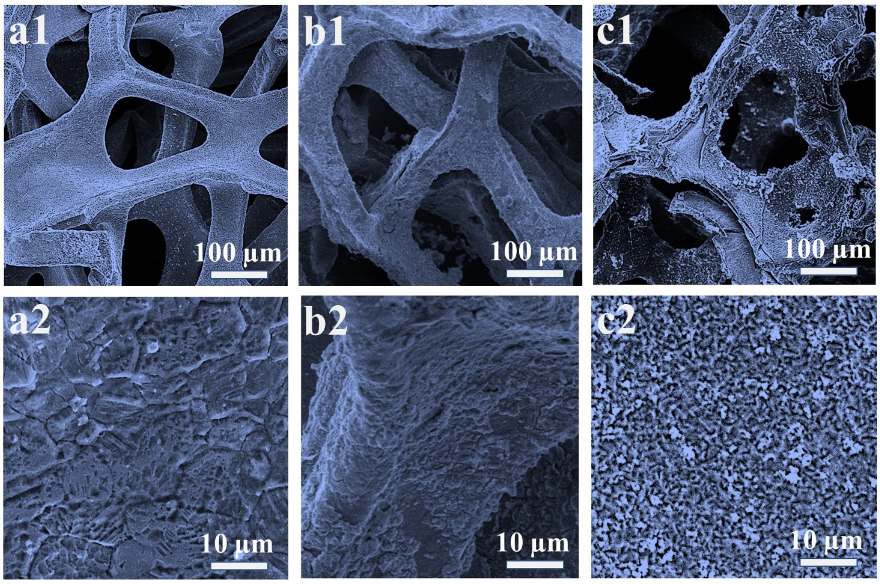


Figure S4. The influence of the amount of thiourea doping on the morphology. (a) Without S; (b) Fe:S=1:1; (c)Fe:S=1:2.

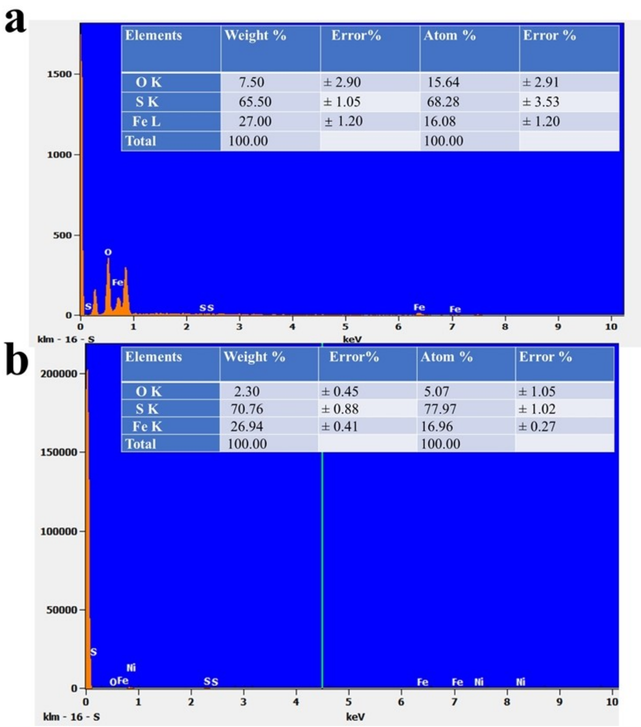


Figure S5. The influence of the amount of thiourea doping on the composition of FeO_x-Ni_3S_2@NF. (a) Fe:S=1:1; (b) Fe:S=1:2.

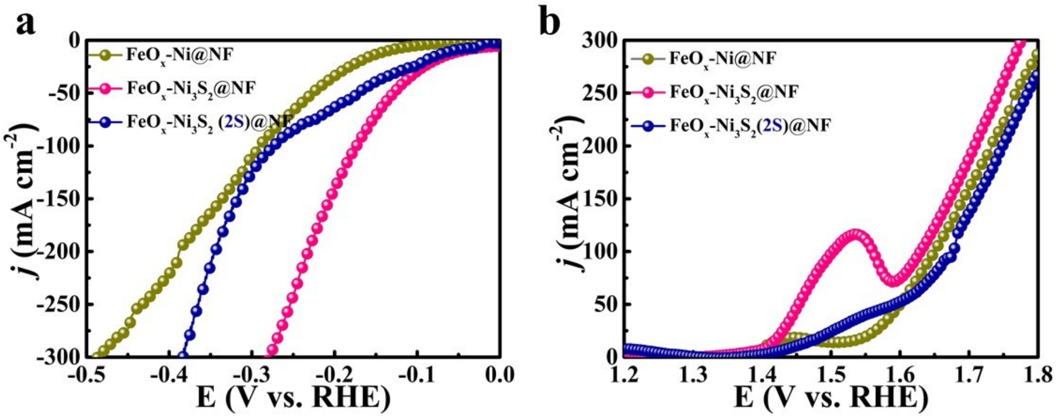


Figure S6. Polarization curves of FeO_x-Ni@NF for HER (a) and OER (b) in 1.0 M KOH with different sulfur source doping amount.

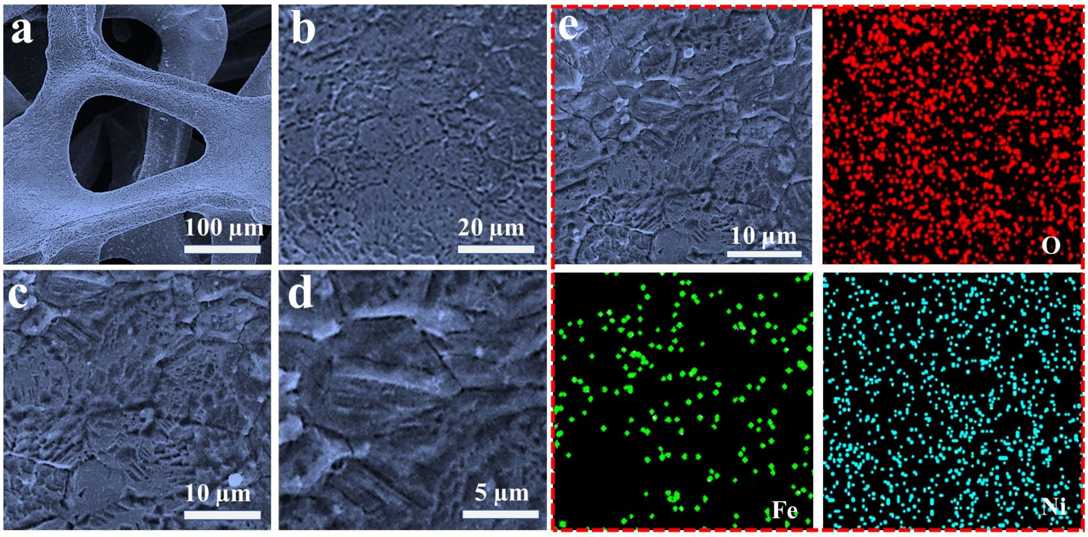


Figure S7. FESEM (a-d) and EDS (e) mapping images of FeO_x-Ni@NF electrode.

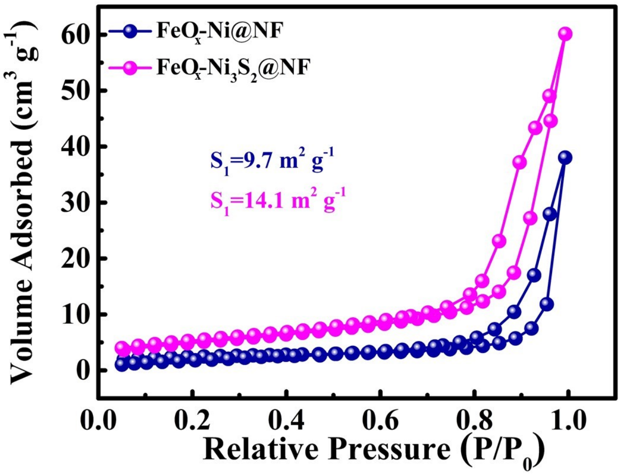


Figure S8. Nitrogen isotherms of the FeO_x-Ni@NF and FeO_x-Ni_3S_2@NF electrodes in nitrogen.

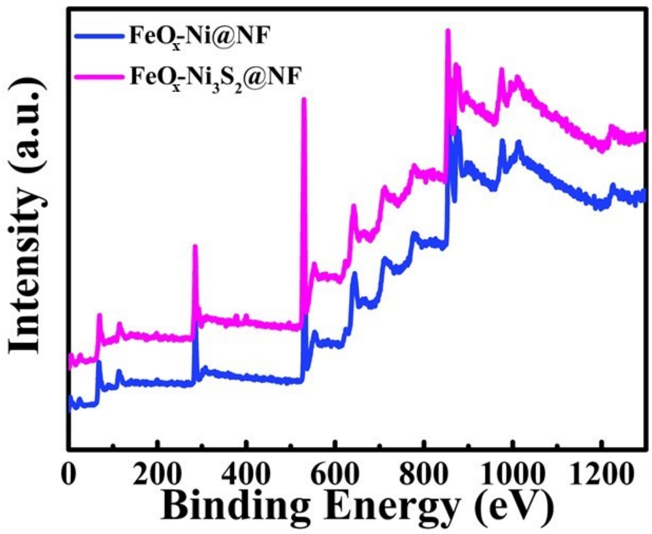


Figure S9. The survey XPS spectrum of the FeO_x-Ni@NF and FeO_x-Ni_3S_2@NF composite.

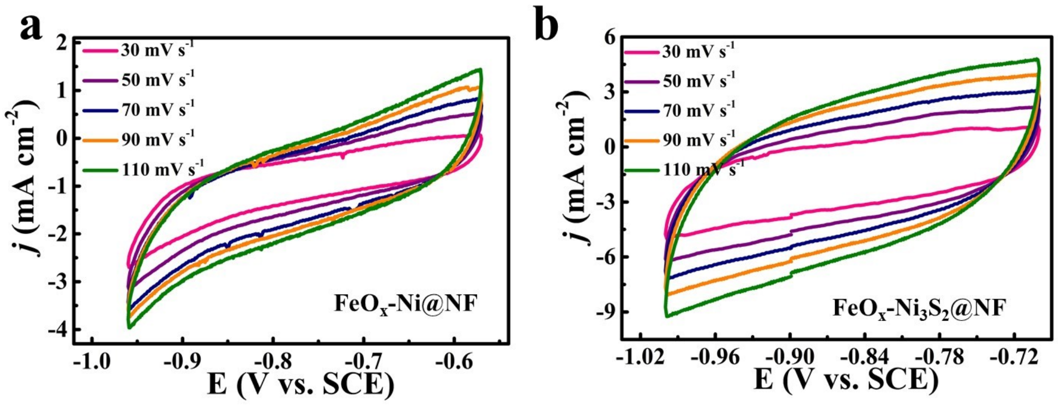


Figure S10. Cyclic voltammograms of (a) FeO_x-Ni@NF and (b) FeO_x-Ni_3S_2@NF electrode in the non-Faradaic current range at scan rates of 10, 30, 50, 70 and 90 mV s^{-1} for HER.

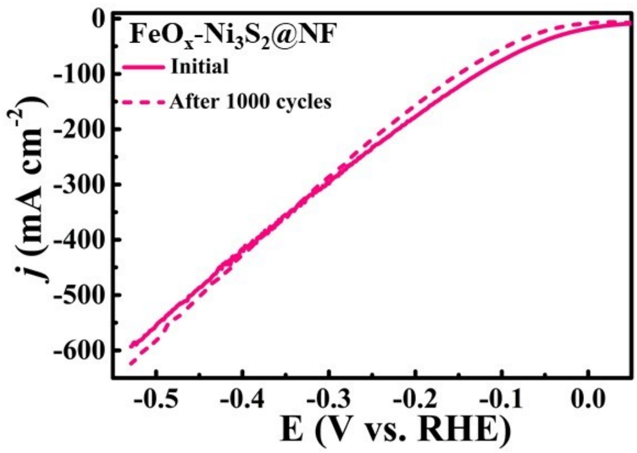


Figure S11. The LSV curves of FeO_x-Ni_3S_2@NF electrode before and after 1000 CV cycles for HER.

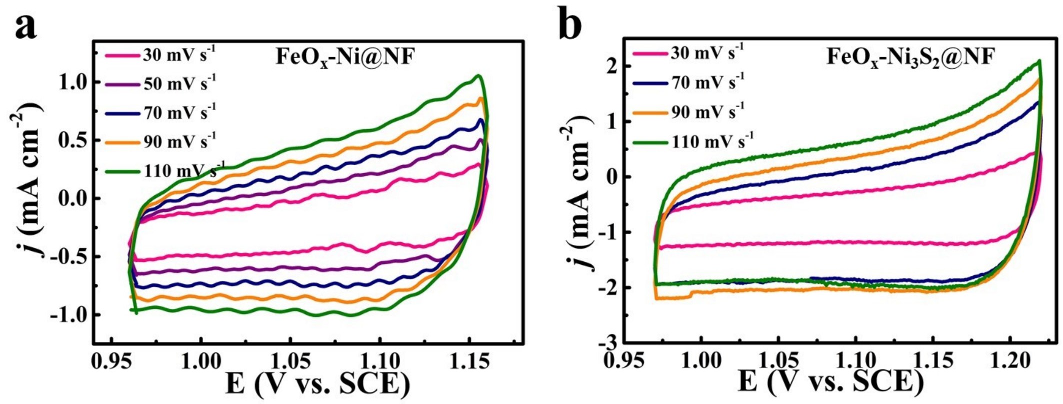
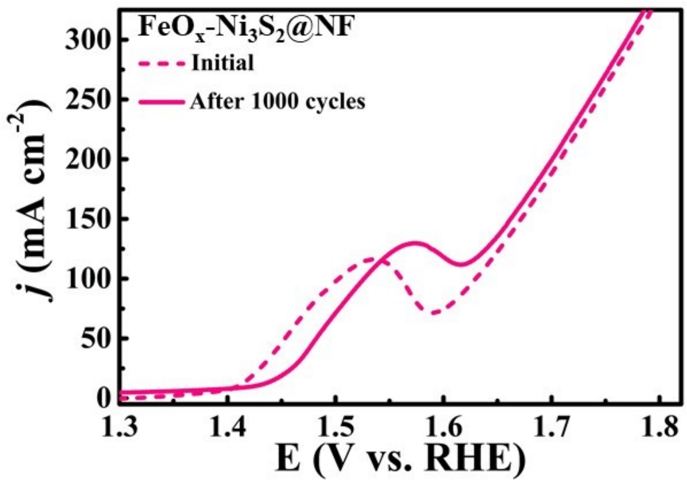


Figure S12. Cyclic voltammograms of (a) FeO_x-Ni@NF and (b) FeO_x-Ni_3S_2@NF electrode in the non-Faradaic current range at scan rates of 30, 50, 70, 90 and 110 mV s^{-1} for OER.



FigureS13. The LSV curves of FeO_x-Ni_3S_2@NF electrode before and after 1000 CV cycles for OER.

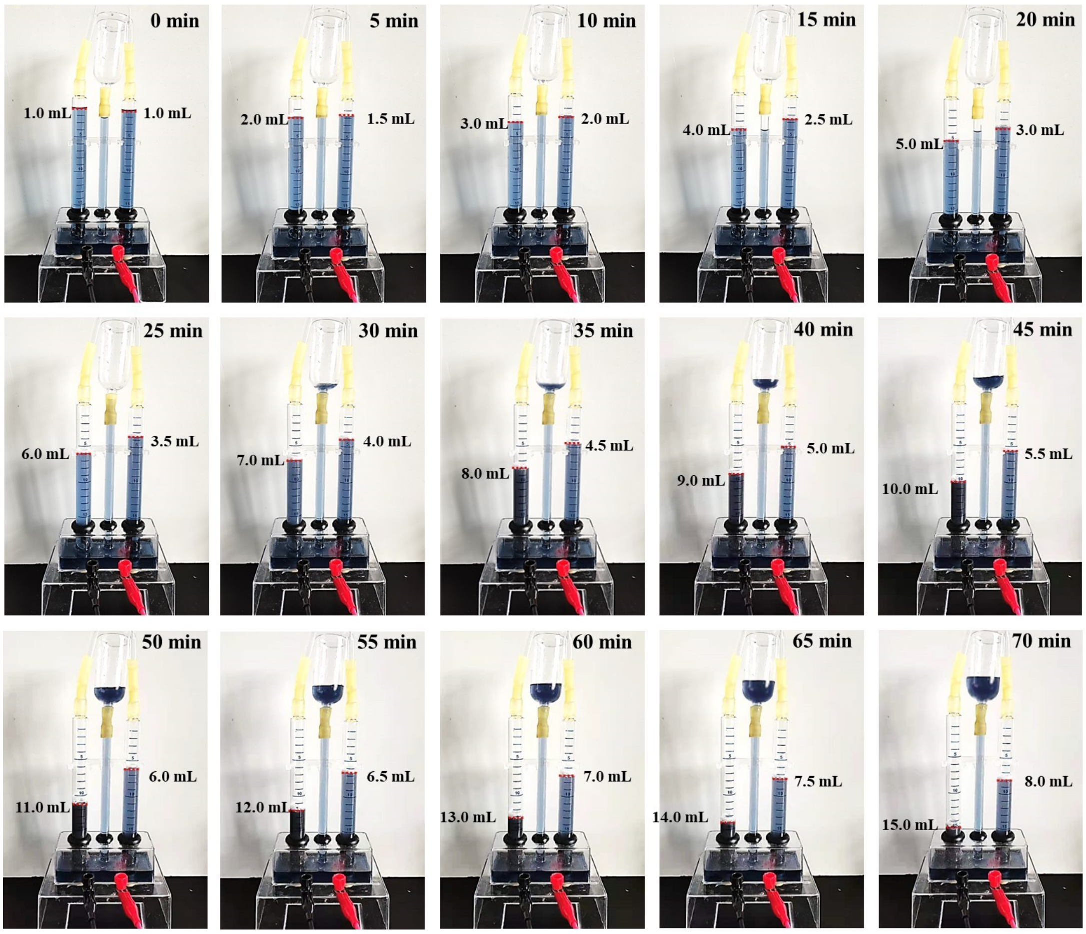


Figure S14. Collection of hydrogen (H_2) and oxygen (O_2) in water splitting in 1.0 M KOH+0.5 M NaCl at the 100 mA cm^{-2}.

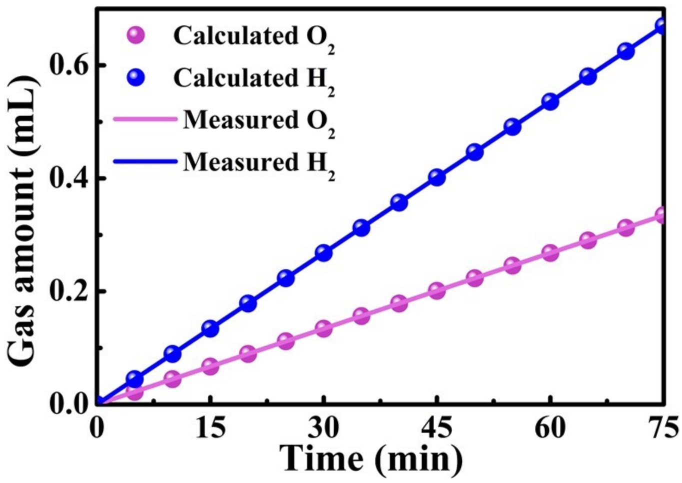


Figure S15. Amount of gas theoretically calculated and experimentally measured versus time for FeO_x-Ni_3S_2@NF || FeO_x-Ni_3S_2@NF.

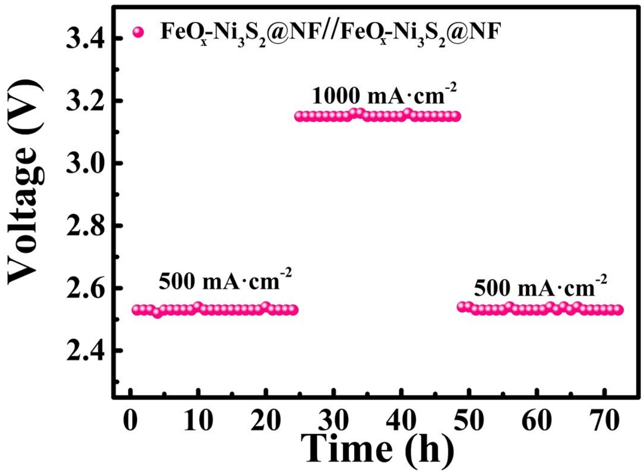


Figure S16. Long-term stability test of FeO_x-Ni_3S_2@NF electrode at current densities of 500 mA cm^{-2}, 1000 mA cm^{-2} and 500 mA cm^{-2} for overall water splitting in 1.0 M KOH+0.5 M NaCl over 72 h.

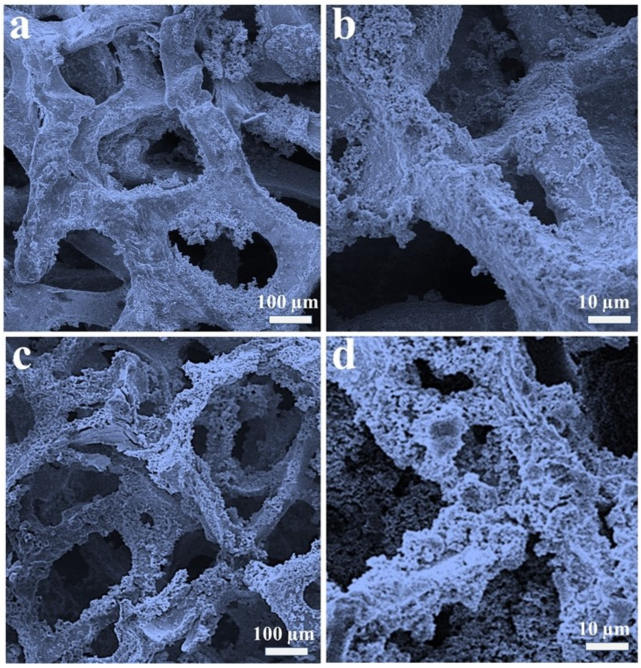


Figure S17. The SEM of FeO_x-Ni_3S_2@NF electrode for HER (a, b) and OER (c, d) at 200 mA cm^{-2} in 1.0 M KOH+0.5 M NaCl after 100 h.

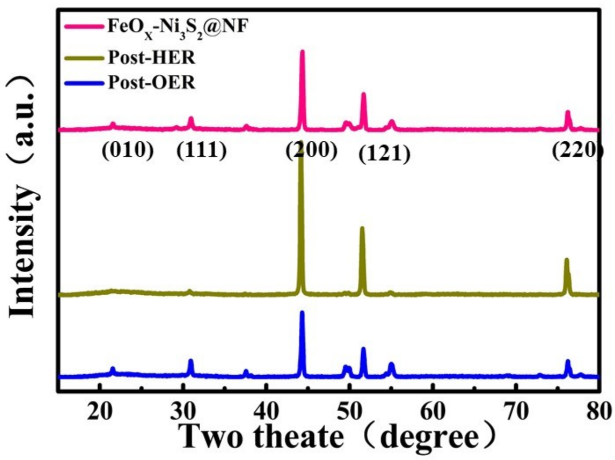


Figure S18. The XRD of FeO_x-Ni_3S_2@NF electrode for HER and OER at 200 mA cm^{-2} in 1.0 M KOH+0.5 M NaCl after 100 h.

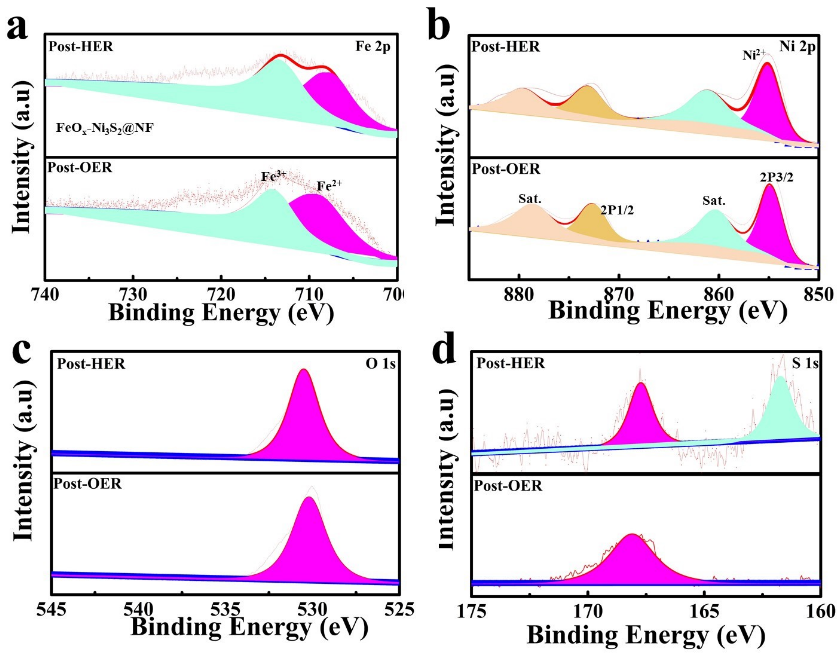


Figure S19. High-resolution XPS of FeO_x-Ni_3S_2@NF electrodes for HER and OER at 200 mA cm^{-2} in 1.0 M KOH+0.5 M NaCl after 100 h. (a) Fe2p; (b) Ni2p; (c) O1s and (d) S1s.

Catalyst	State	Atomic Ratio Fe : S
FeO_x-Ni_3S_2@NF		1:4.9
	Post-HER	1:4.1
	Post-OER	1:3.4

Table S1. ICP-AES analysis of the FeO_x-Ni_3S_2@NF electrode

Table S2. Comparison the HER performance of FeO_x-Ni_3S_2@NF electrode with other electrocatalysts in 1.0 M KOH.

Catalysts	j (mA cm^{-2})		Taf slope	References
	10	100	(mV dec^{-1})	
FeO_x-Ni_3S_2@NF	**52**	**178**	**57**	**This Work**
Ni Foam/P-$CoMoO_4$-350	94	197	93	Adv. Sci. **2020**, 7, 1903674
H-MoS2/MoP	92	-	59.8	Small **2020**, 16, 2002482
NiO/Ru@PNS	39	-	75	J. Mater. chem. A, **2019**, 7, 2344-2350
H-NiFe LDH	59	-	62.3	Energy Environ. Sci., **2019**, 12, 572-581
Pt/np-$Co_{0.85}$Se	55	-	35	Nature Communications, **2019**, 10, 1743
MoS_2-MoP/NC	35	69	30	Nano Energy, **2020**,78, 105253
Co, Mo_2C-CNF	128	-	60	Chemical Engineering Journal **2020**, 125481
MoP/0.5CM-CDs1100	70	-	77.49	Nano Energy **2020**, 72, 104730
CC@N-CoP	42	-	41.2	Adv. Mater. **2018**, 1800140
Mo_2N /CeO_2@NF	26	-	37.8	ACS Appl. Mater. Interfaces **2020**, 12, 26, 29153-29161
b-S-Ni_3Se_4Se-Ni_3S_2/NF	89	-	61	Nano Energy **2020**, 74, 104787
Ni_2P–$Ni_{12}P_5$/ NF	76	147	68	Small **2020**, 06770
VS_2-Mo-10	243	-	52.6	Chemical Engineering Journal **2020**, 396 125227
$Re_{1-x}Mo_xSe_2$	77	-	42	ACS Nano **2020**, 14, 9, 11995–12005
Mo_6Te_6/$MoS_{2(1-x)}$ Te_{2x}	320	-	55.7	Small **2020**, 2004296
N-LDH/2D-Pt	31	-	32.3	ACS Nano **2020**, 14, 8, 10578–10588
Ru SAs–Ni_2P	57	-	75	Nano Energy **2020**,105467
Fe-Ni_3S_2 @$FeNi_3$	105	-	69	Chemical Engineering Journal **2020**, 396, 125315
$(Ni_{0.75}Fe_{0.25})_2P$@GCs	83	-	70.4	J. Mater. Chem. A, **2019**,7, 20357-20368
CoNi/$CoFe_2O_4$/NF	82	189	45	J. Mater. Chem. A, **2018**,6, 19221-19230
NiO/Ru@PNS	39	-	75	J. Mater. Chem. A, **2019**,7, 2344-2350

Table S3. Comparison the OER performance of FeO_x-Ni_3S_2@NF electrode with other electrocatalysts in 1.0 M KOH.

Catalysts	OER (10 mA cm^{-2})	Tafel Slope (mV dec^{-1})	References
FeO_x-Ni_3S_2@NF	**196**	**62**	**This work**
CoNi-OH	270	73.5	Chem. Eng. J. **2020**, 401,126092
Ru-HPC	310	60.7	Nano Energy **2019**, 58, 1-10
$FeNi_3S_2$/NF	214	42	ACS Catal. **2018**, 8, 5431-5441
NiFeMo	238	35	ACS Energy Lett. **2018**, 3, 546-554
NiCoP@NC NA/NF	215	70.5	Adv. Funct. Mater. **2019**, 29, 1906316
δ-FeOOH NSs/NF	265	36	Adv. Mater. **2018**, 30, 1803144
Ni_2P-VP_2	220	49	Adv. Mater. **2019**, 31, 1901174
Ni/Ni$(OH)_2$@NF	270	53	Adv. Mater. **2020**, 32, 1906915
Ni–Fe–Mo/NF	255	35	Adv. Sci. **2020**, 7, 1902034
Ni-ZIF/Ni-B@NF	234	76	Adv. Energy Mater. **2020**, 10, 1902714
Ni/FeOOH@NF	239	70.5	J. Mater. Chem. A, **2020**, 8, 12603–12612
YP-Co(OH)F	238	67	Small **2019**, 15, 1904105
Co6W6C@NC	286	53.96	Small **2020**, 16, 1907556
Ni–Ni3C/CC	299	43.8	Small **2020**, 16, 2001642
FexNi$_{3-x}$S2@NF	252	64	Adv. Energy Mater. **2020**, 10, 2001963
NiFe LDH	187	34.42	Energy Environ. Sci., **2019**, 12, 572-581
2D NiCoFe/NF	240	58	Nanoscale, **2018**, 10, 12975–12980

Table S4. The overall-water splitting performance for FeO_x-Ni_3S_2@NF and other electrodes with NF-base electrocatalysts in 1.0 M KOH.

Catalysts	Current Density ($mA\ cm^{-2}$) 10	100	References
FeO_x-Ni_3S_2@NF	**1.41**	**1.61**	**This Work**
CoNi/$CoFe_2O_4$/Ni	1.57	1.75	J. Mater. Chem. A, **2018**, 6,19221
MoS_2/Co_9S_8/Ni_3S_2/Ni	1.54		J. Am. Chem. Soc. **2019**, 141, 10417−10430
NiFeMo/NF	1.45		ACS Energy Lett. **2018**, 3, 546−554
$FePO_4$/NF	1.54	1.72	Adv. Mater. **2017**, 29, 1704574
Ni-$NiOH_2$/NF	1.59		Adv. Mater. **2020**, 32, 1906915
NiFeMoOOH/NF	1.5	1.63	Adv. Sci. **2020**, 7, 1902034
Ni-ZIF/Ni-B@NF	1.54		Adv. Energy Mater. **2020**, 10, 1902714
NiP_2/Ni_2P@Ni	1.54		J. Am. Chem. Soc. **2019**, 141, 10417−10430
YP-Co(OH)F	1.54		Small **2019**, 15, 1904105
Co_6W_6C@NC	1.585		Small **2020**, 16, 1907556
Ni–Ni_3C/CC	1.64		Small **2020**, 16, 2001642
Ni/$Ni(OH)_2$/NF	1.59		Adv. Mater. 2020, 32, 1906915
NiFe-Mo/OOH@NF	1.5	1.63	Adv. Sci. **2020**, 7, 1902034
h-NiS_x/NF	1.54	1.72	Adv. Mater. **2017**, 29, 1704574
δ-FeOOH NSs/NF	1.62		Adv. Mater. **2018**, 30, 1803144
$NixCo_{2-x}P$@NC	1.56		Adv. Funct. Mater. **2019**, 29, 1906316
Fe-Ni_3S_2/NF	1.54		ACS Catal. **2018**, 8, 5431−5441
Ni3N-VN/NF	1.51		Adv. Mater. **2019**, 31, 1901174
Ni/Mo_2C(1:2)-NCNFs	1.64		Adv. Energy Mater. **2019**, 9, 1803185