Supporting Information

Hierarchical Co(OH)F/CoFe-LDH heterojunction enabling high-performance overall water-splitting

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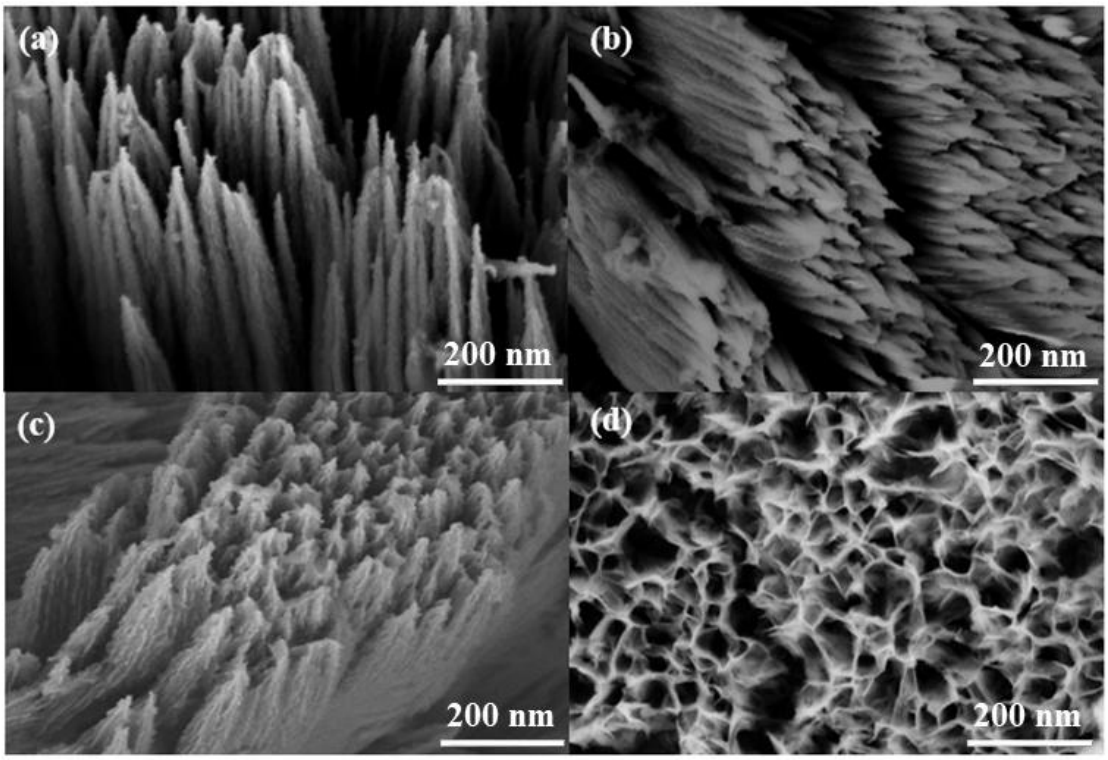


Figure. S1. SEM images of CoFe-LDH on Co(OH)F under different electrodeposition times. (a) 10 s; (b) 20 s; (c) 40 s; (d) 60 s.

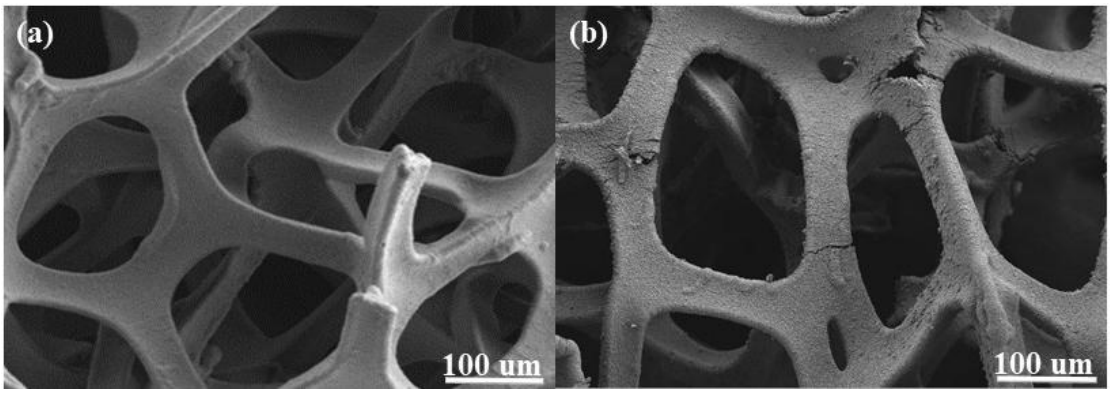


Figure. S2. (a) SEM image of bare NF after removing the oxide layer with 3 M HCl treatment. (b) SEM image of NF surface covered by Co(OH)F @CoFe-LDH.

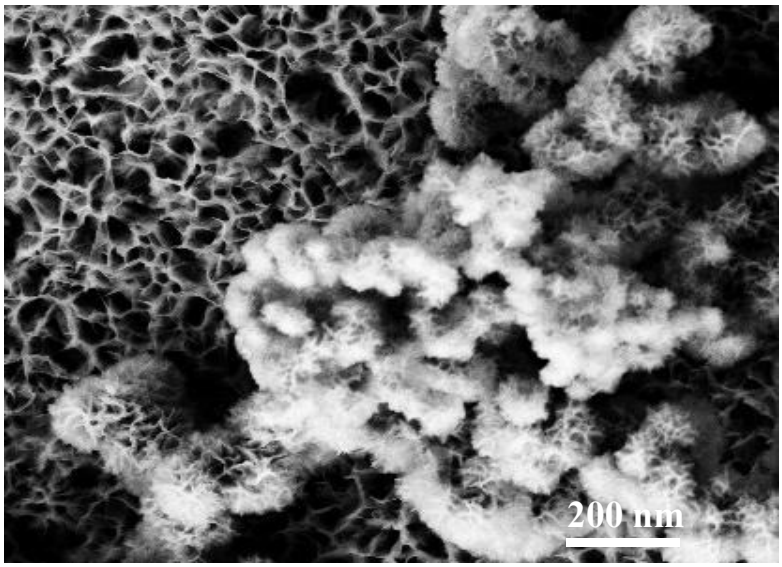
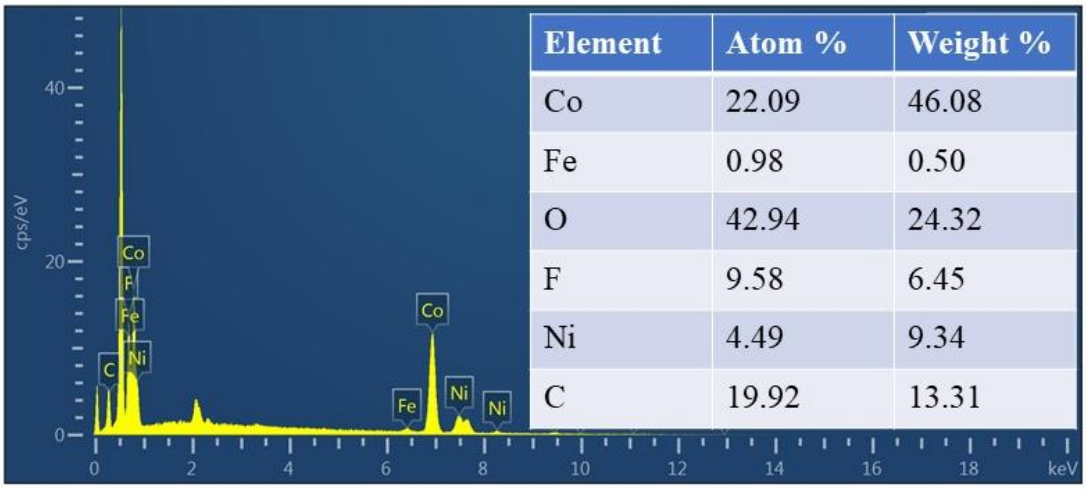


Figure. S3. SEM image of the CoFe-LDH



Element	Atom %	Weight %
Co	22.09	46.08
Fe	0.98	0.50
O	42.94	24.32
F	9.58	6.45
Ni	4.49	9.34
C	19.92	13.31

Figure. S4. EDS spectrum of Co(OH)F@CoFe-LDH.

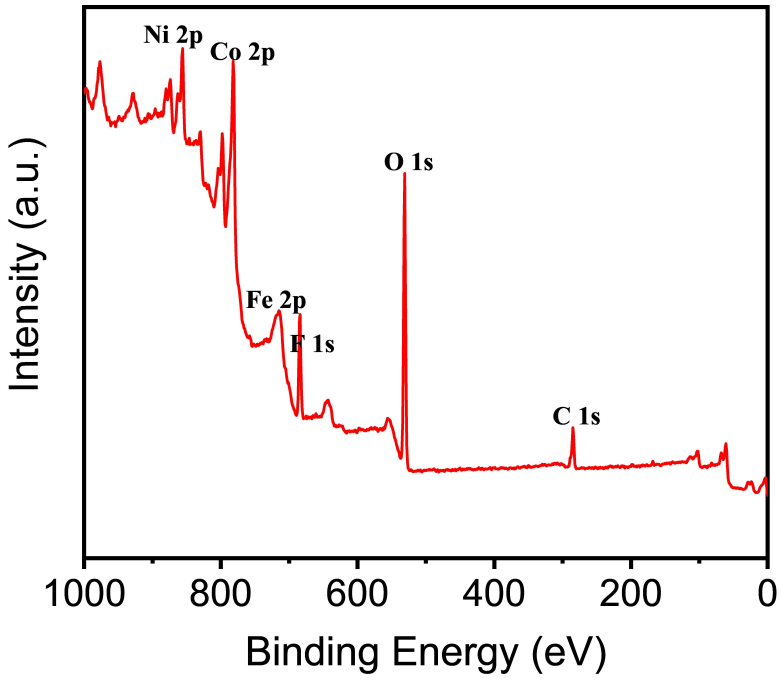


Figure. S5. XPS survey spectrum of Co(OH)F@CoFe-LDH

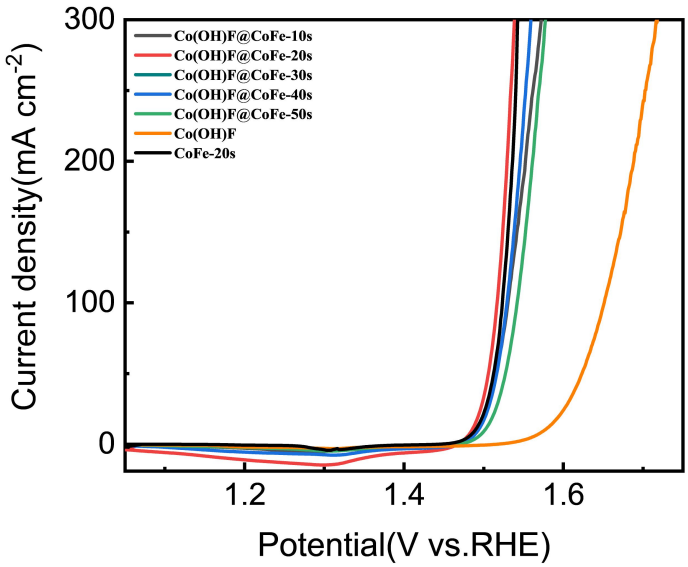


Figure. S6. Polarization curves of different Co(OH)F@CoFe-LDH samples prepared with different electrodeposition times for OER.

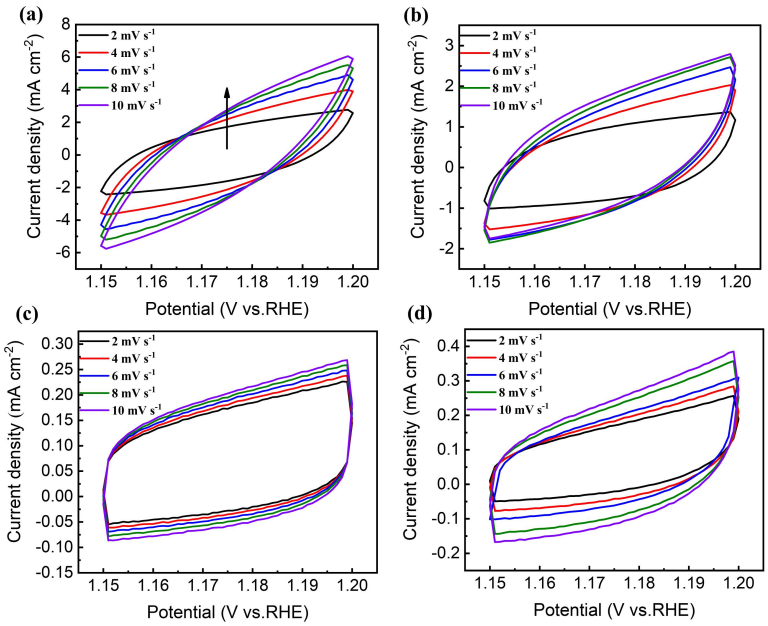


Figure. S7. CV curves of the catalysts recorded in the region of 1.15-1.20 V. (a) Co(OH)F@CoFe-LDH, (b) Co(OH)F, (c) NF, (d) CoFe-LDH.

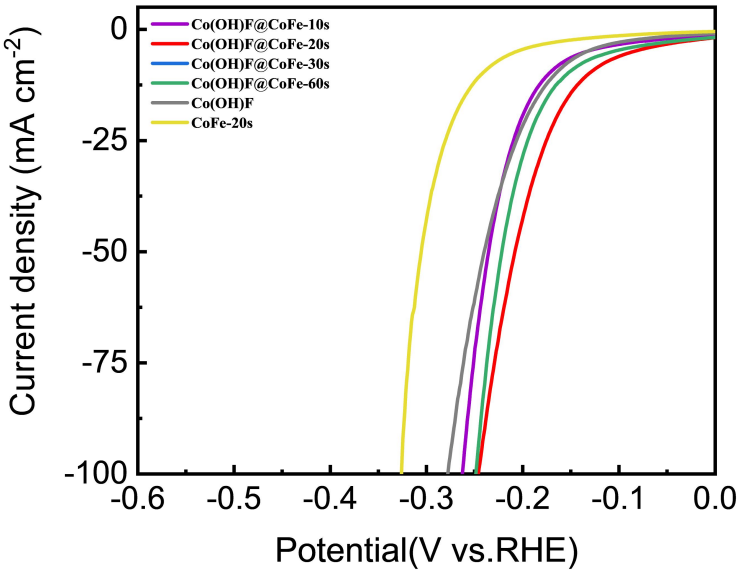


Figure. S8. Polarization curves of different Co(OH)F@CoFe-LDH samples prepared with different electrodeposition times for HER

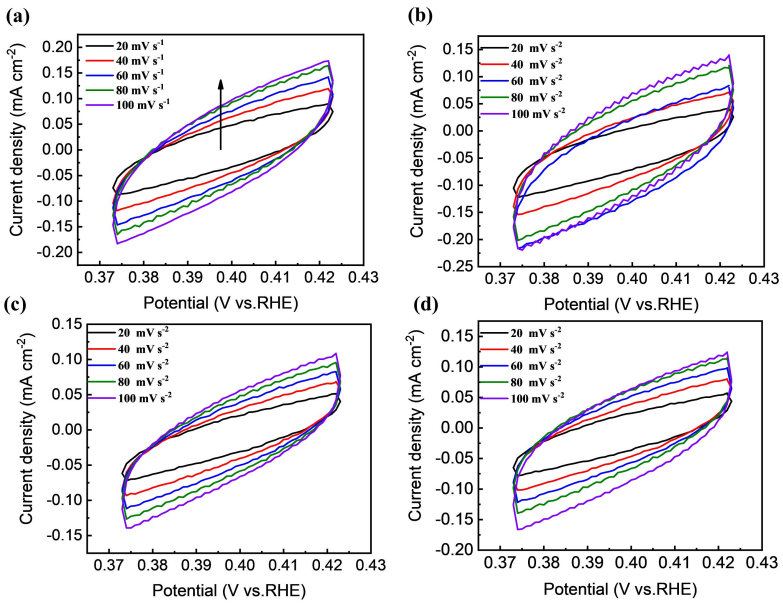


Figure. S9. CV curves of the catalysts recorded in the region of 0.37-0.42 V. (a) Co(OH)F@CoFe-LDH, (b) Co(OH)F, (c) NF, (d) CoFe-LDH.

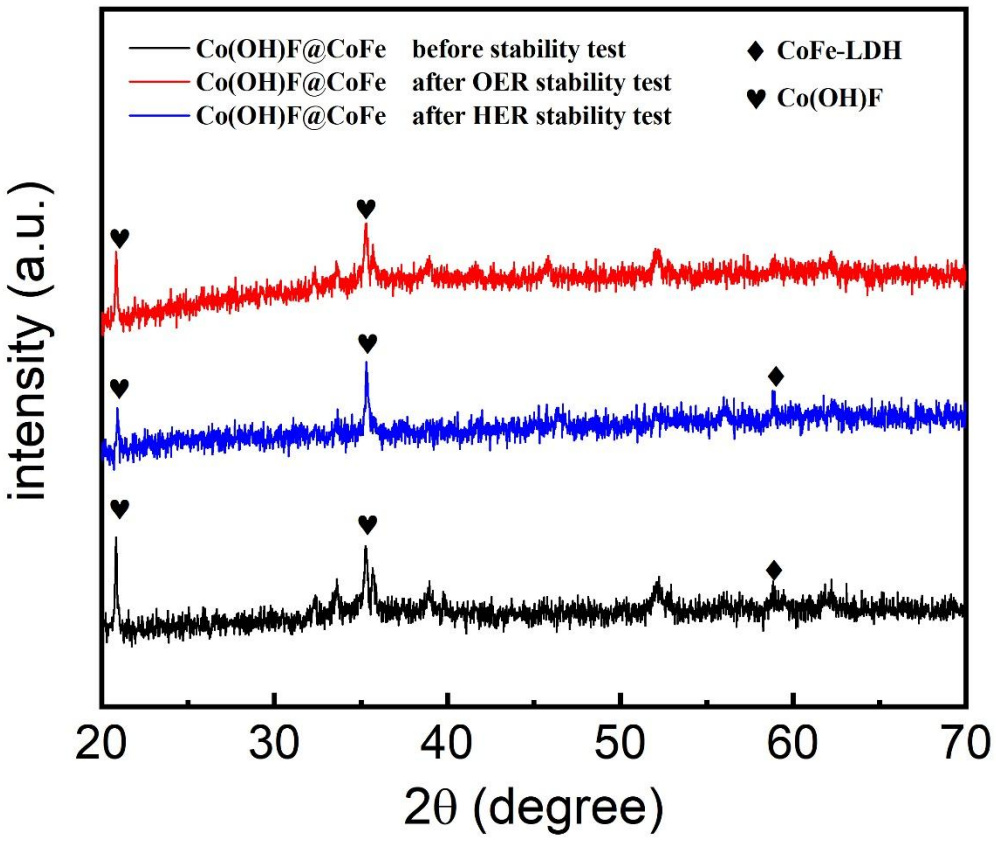


Figure. S10. XRD spectra of the Co(OH)F@CoFe-LDH before and after OER and HER stability tests

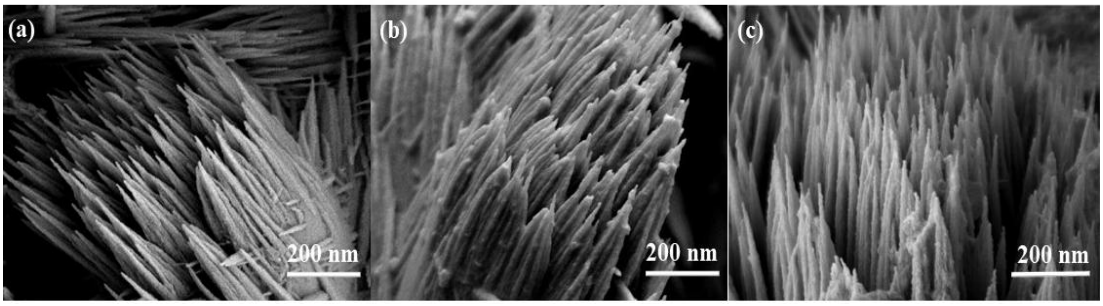


Figure. S11. (a) SEM images of Co(OH)F@CoFe-LDH before stability tests. (b) SEM images of Co(OH)F@CoFe-LDH after stability tests (OER). (c) SEM images of Co(OH)F@CoFe-LDH after stability tests (HER)

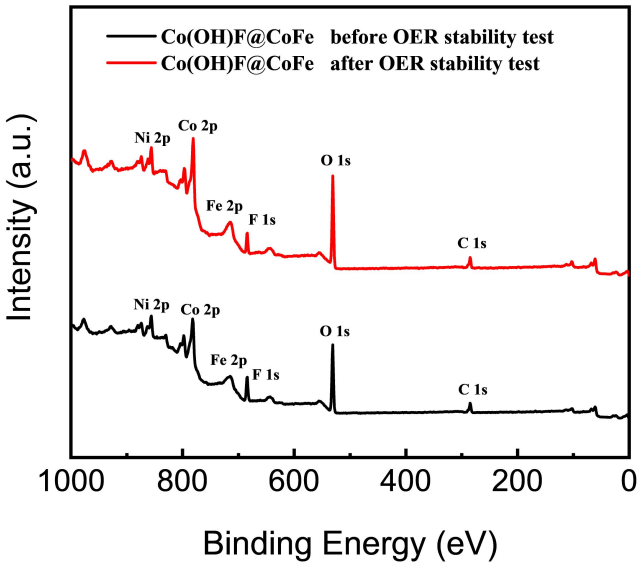


Figure. S12. XPS survey of Co(OH)F@CoFe-LDH before and after stability tests

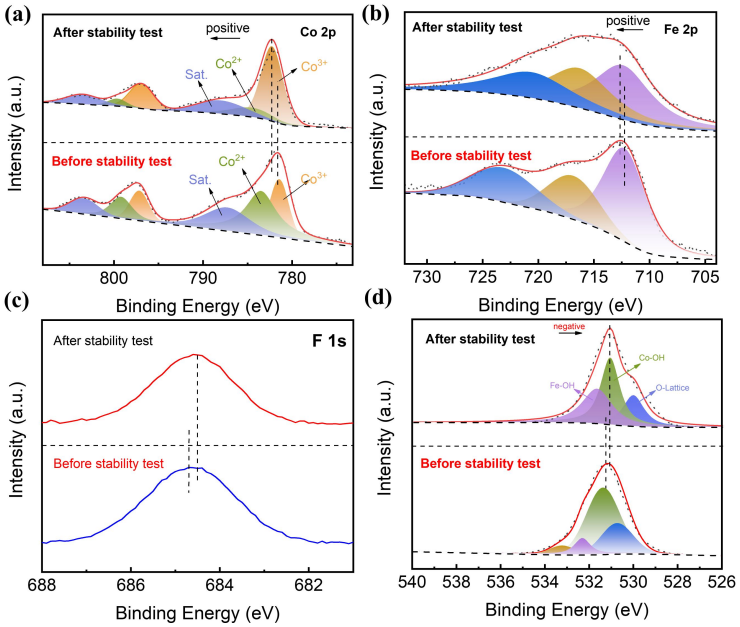


Figure. S13. high-resolution XPS spectra of (a) Co 2p, (b) Fe 2p, (c) F 1s and (d) O 1s of Co(OH)F@CoFe-LDH before and after stability tests.

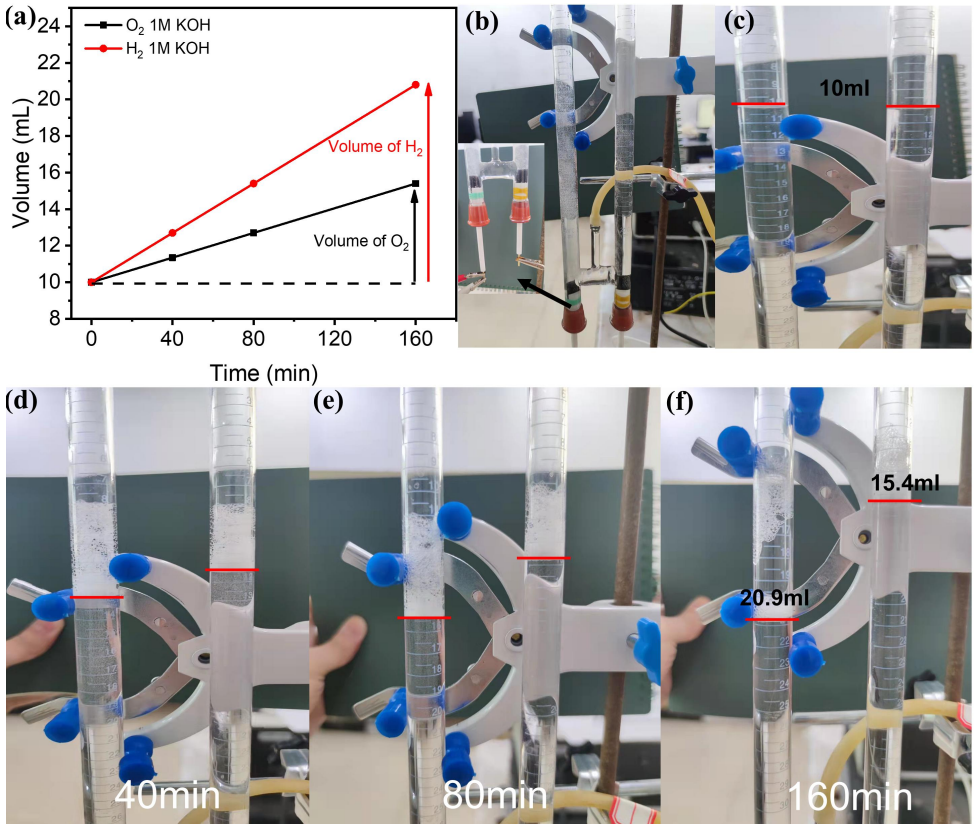


Fig. S14. (a) Diagram of the amount of H_2 and O_2 released over time in 1 M KOH. (b) Device diagram for measuring Faraday efficiency. (c-f) Corresponding levels of H_2 and O_2 gases generated at different times 1 M KOH electrolyzer.

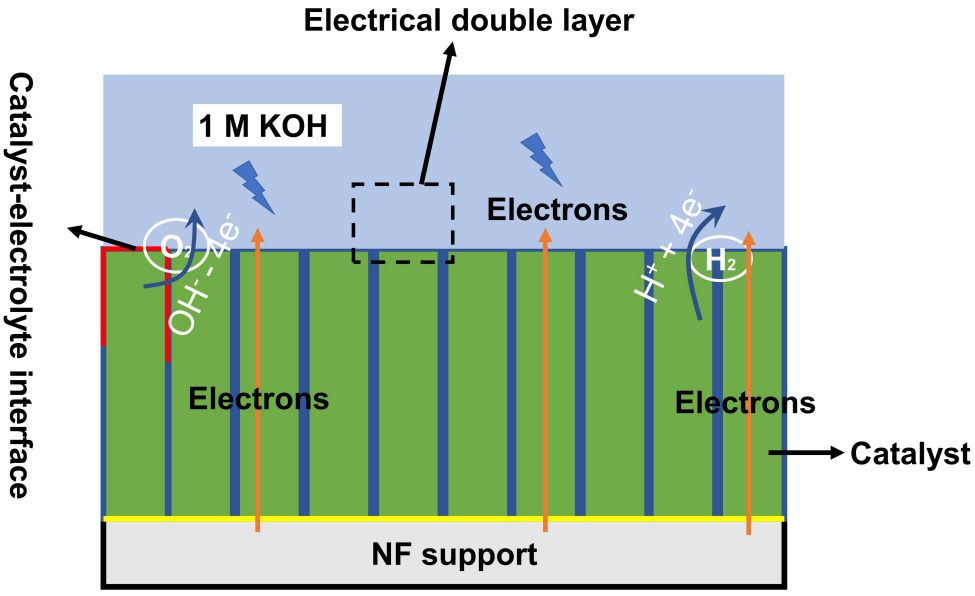


Fig. S15. Schematic illustration of the electrocatalytic HER and OER in 1M KOH solution.

Table S1. ICP-MS results of catalysts

Catalysts	ICP-MS (wt %)		
	Co	Fe	Co: Fe (at.)
Co(OH)F@CoFe-LDH	51.29	1.2	42.7:1
CoFe-LDH	23.58	6.7	3.4:1

Table. S2. Electrocatalytic activity of OER, HER and overall water splitting for the reported various electrocatalysts in 1 M KOH at the current density of j mA/cm^2.

Catalysts	HER		OER		Overall water splitting	References
	η (mV) @j (mA/cm^2)	Tafel Slope (mV/dec)	η (mV) @j (mA/cm^2)	Tafel Slope (mV/dec)	η (V) @j (mA/cm^2)	
Co(OH)F@CoFe-LDH/NF	130@10	82.9	240@10	25.4	1.58@10	This work
	208@50	82.9	262@50	25.4	1.67@50	
	245@100	82.9	274@100	25.4	1.7@100	
CoFe@NiFe-200/NF	240@10	84.69	190@10	45.71	1.59@10	[1]
NiFe LDH-NiSe/NF	276@100	70	240@100	65.6	1.53@10	[2]
$NiCo_2S_4$@NiFe-LDH	200@10	46.3	201@60	101.1	1.6@10	[3]
Co_3S_4@MoS_2	210@10	88	330@10	59	---	[4]
Ni_3S_2-$CoMoS_x$/NF	234@10	125	90@10	75	1.49@10	[5]
FeS/NiS/NF	144@10	120	203@10	39	1.618@10	[6]
Cu@CoFe-LDH	171@10	36.4	240@10	44.4	1.681@10	[7]
NiFe-LDH-Co_3O_4	162@10	105	214@10	30	1.64@10	[8]
$NiCo_2O_4$@NiO@Ni	124@10	58	240@10	43	1.6@10	[9]
CoS-$Co(OH)_2$ @aMoS_{2+x}	143@10	68	380@10	68	1.58@10	[10]

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