

Supporting Information

Enhanced electrocatalytic activity of layered triple hydroxide (LTH) by modulating electronic structure and active sites for efficient and stable urea electrolysis

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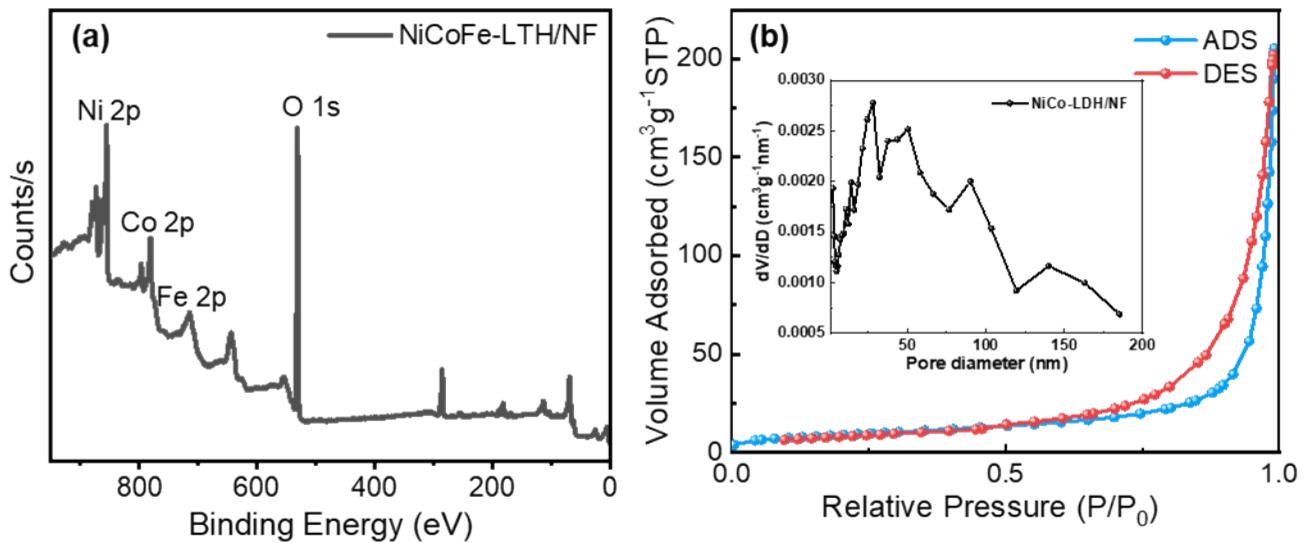


Figure S1. (a) XPS survey spectrum of NiCoFe-LTH/NF, (b) N₂ adsorption and desorption isotherms and pore size distribution of NiCo-LDH/NF.

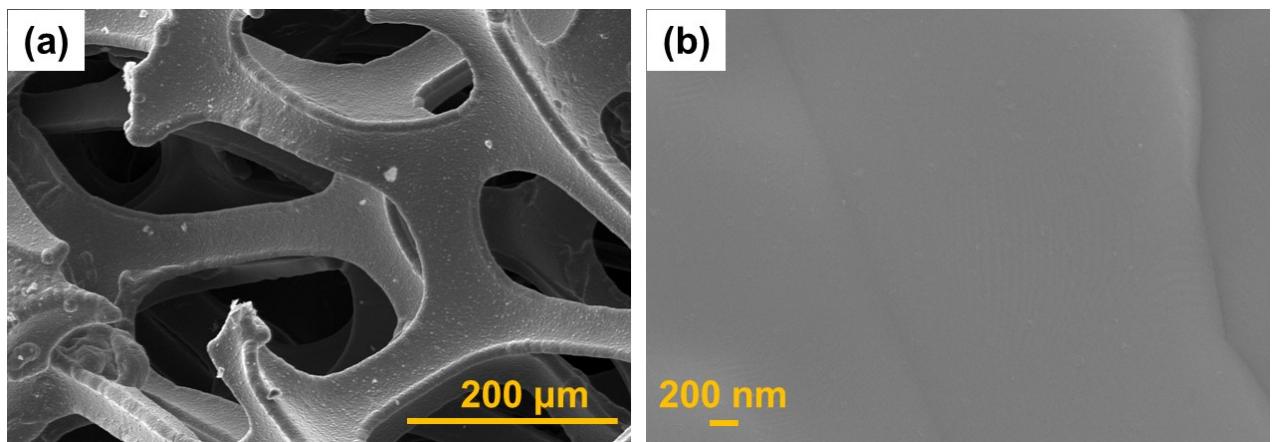


Figure S2. (a-b) SEM images of Blank NF.

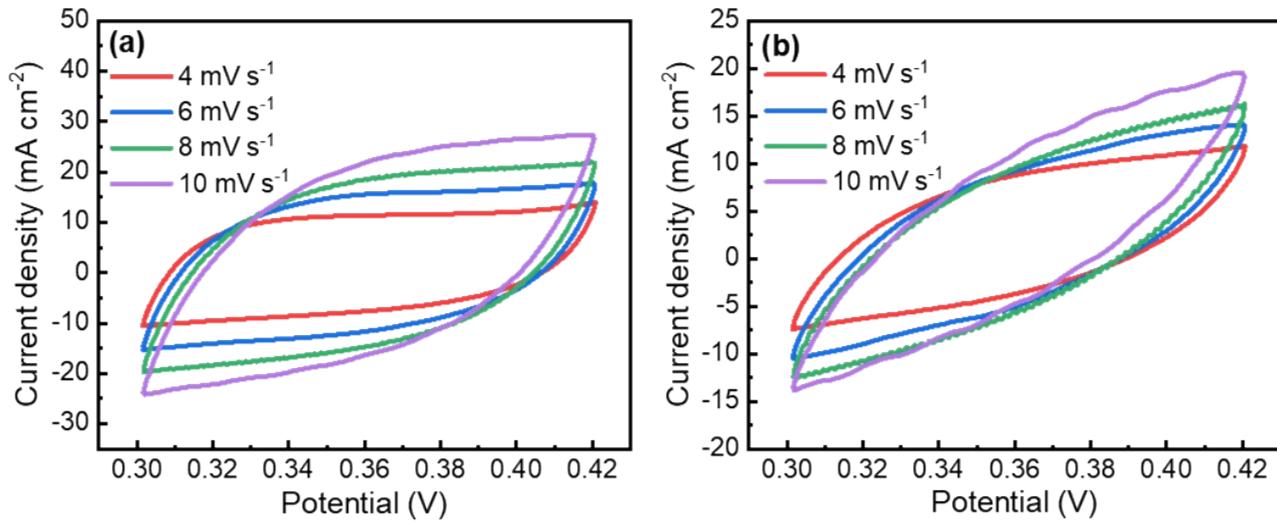


Figure S3. CV curves of (a) NiCoFe-LTH/NF and (b) NiCo-LDH/NF at different scan rates.

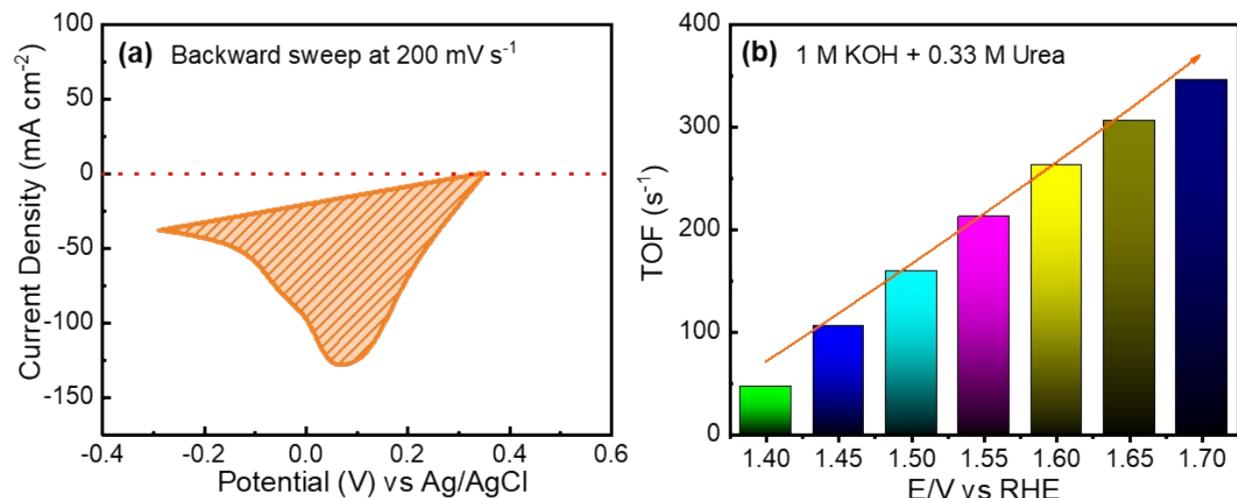


Figure S4. (a) Backward CV of NiCo-LDH/NF for charge integration and calculation of electrochemically accessible sites, (e) Plot of TOF against overpotential for UOR (0.33 M urea in 1 M KOH) for NiCo-LDH/NF.

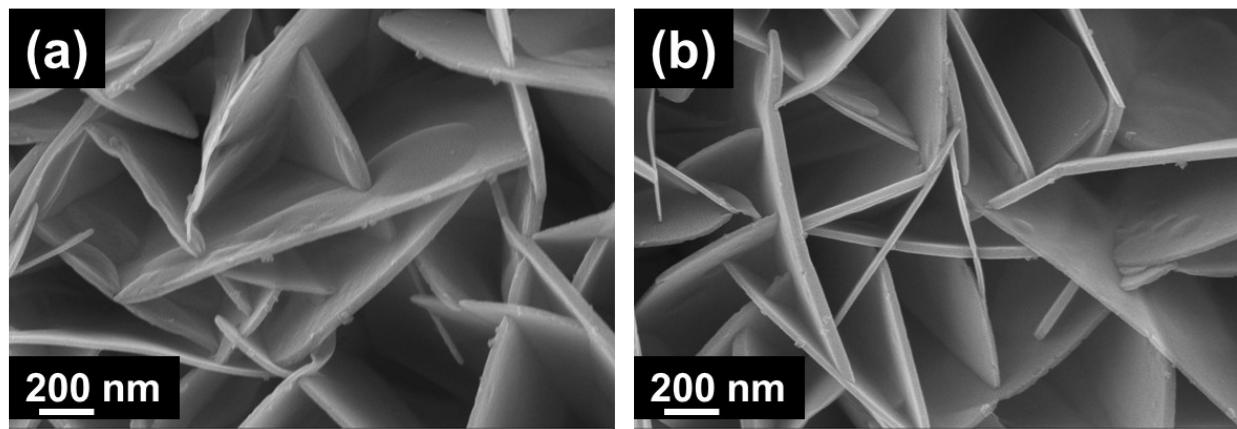


Figure S5. FE-SEM images of NiCoFe-LTH/NF nanosheets after 30 h stability for (a) UOR and (b) HER.

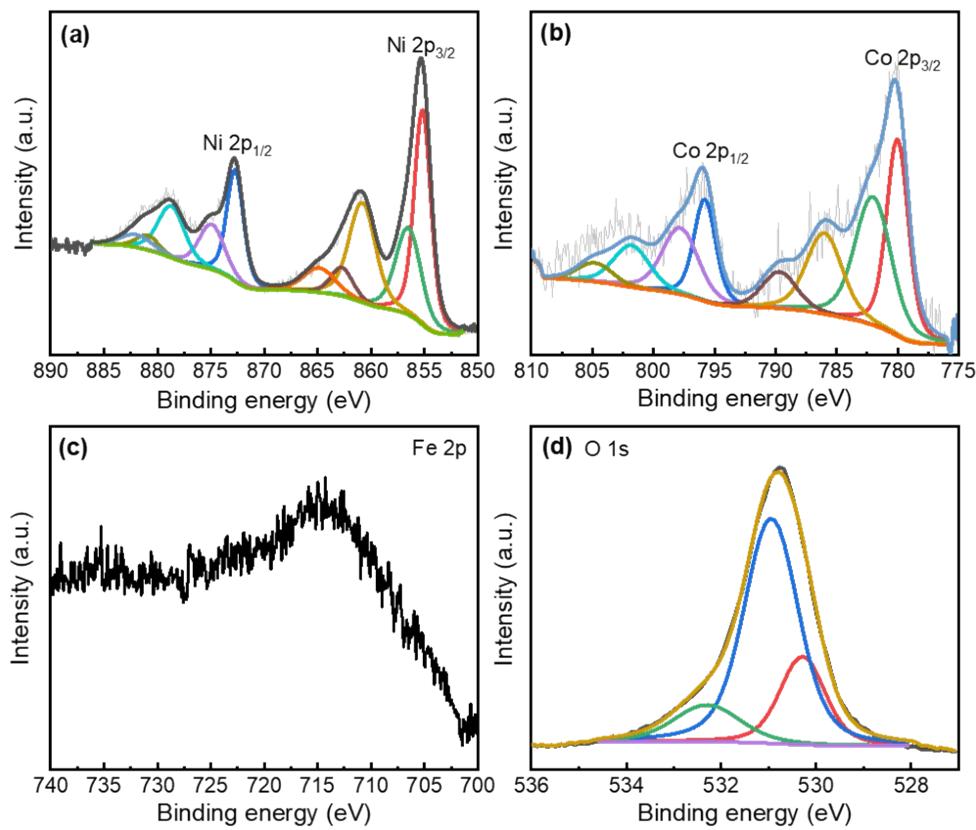


Figure S6. XPS spectra of (a) Ni 2p, (b) Co 2p, (c) Fe 2p, and (d) O 1s of NiCoFe-LTH/NF after long term UOR stability.

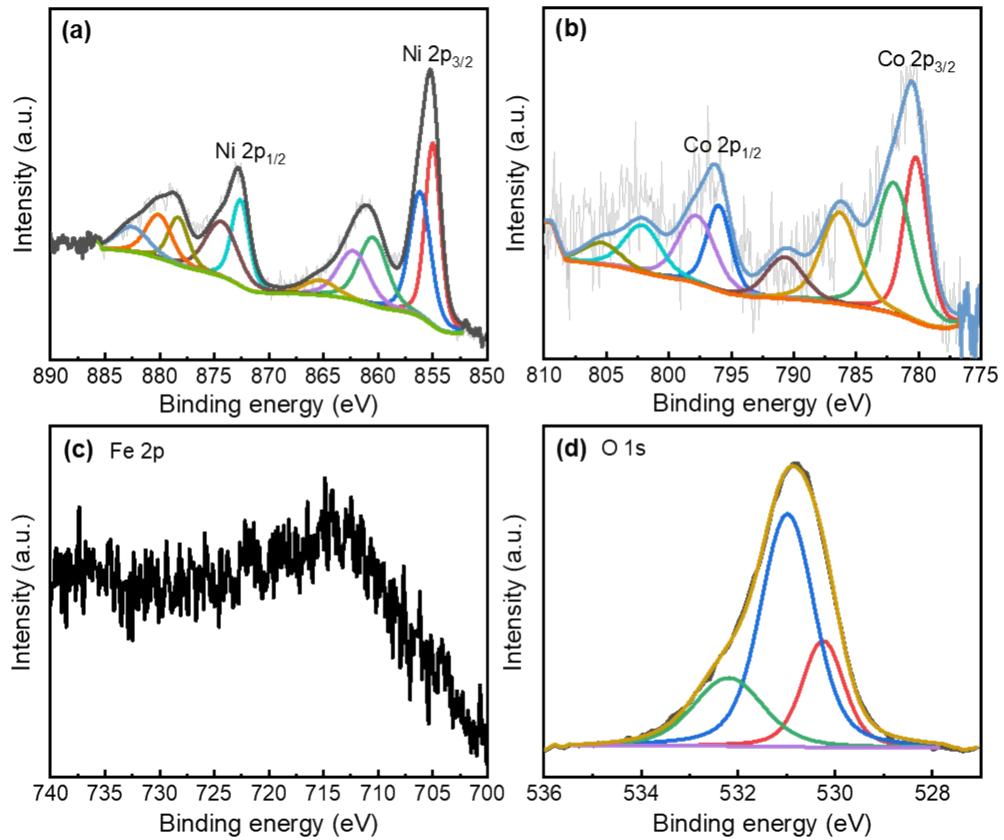


Figure S7. XPS spectra of (a) Ni 2p, (b) Co 2p, (c) Fe 2p, and (d) O 1s of NiCoFe-LTH/NF after long term HER stability.

Table S1. Comparison with other electrocatalysts for UOR reported in literatures

Electrocatalyst	Concentration of urea (M)	Current Density (mA cm ⁻²)	Potential (V)	References
Ni(OH) ₂ on Au substrate	5M KOH/1M urea	10	1.37 (vs RHE)	Electrochim. Acta 108 (2013) 660-665
Ni ₃ N/Carbon cloth (array)	0.33	100	0.462 (vs SCE)	Inorg. Chem. Front., 2017, 4, 1120-1124
Ni foil	0.33	10	1.51 (vs RHE)	J. Power Sources 196 (2011) 9579-9584
Ni ₂ P/Carbon cloth (array)	0.5	100	0.447 (vs SCE)	J. Mater. Chem. A, 2017, 5, 3208-3213
Ni(OH) ₂ -nanocup arrays	0.33	10	1.46 (vs RHE)	Electrochim. Acta 144 (2014) 194-199
Ni(OH) ₂ nanosheets	0.33	10	0.452 (vs SCE)	Angew. Chem. Int. Ed., 2016, 55, 12465-12469
Ni/Co	0.33	10	1.45 (vs RHE)	Sci Rep. 4 (2014) 5863
MnO ₂ nanosheets	0.5	100	0.445 (vs SCE)	Angew. Chem. Int. Ed., 2016, 55, 3804-3808
α -Ni(OH) ₂ nanotubes	0.33	10	1.60 (vs RHE)	Electrochim. Commun. 29 (2013) 21-24
Ni(OH) ₂ (array)	0.33	10	0.312 (vs	Electrochim. Acta, 2014, 144, 194-199

			SCE)	
Metallic Ni(OH) ₂	0.33	10	1.40 (vs RHE)	Angew. Chem. Int. Ed. Engl. 55 (2016) 12465-12469
NiO/Ni foam (array)	0.33	10	0.312 (vs SCE)	J. Power Sources, 2014, 272, 711-718
NiMo/graphene	0.33	10	1.40 (vs RHE)	Electrochim. Acta 242 (2017) 247- 259
NiFe double hydroxide	0.33	10	1.48 (vs RHE)	New J. Chem. 41 (2017) 4190-4196
NiCo ₂ O ₄	0.33	10	1.36 (vs RHE)	Nanoscale 6 (2014) 1369-1376
CuO-Ni(OH) ₂ nanosheets	0.33	10	1.41 (vs RHE)	Applied Surface Science 560 (2021) 150009
			1.482	
NiCo-LDH/NF	0.33	25	(vs RHE)	This work
			1.337	
NiCoFe- LTH/NF	0.33	25	(vs RHE)	This work

Table S2. Comparison with other electrocatalysts for HER reported in literatures

Electrocatalyst	Electrolyte	Overpotential (mV) (10 mA cm ⁻²)	References
Ni nanowires	1 M NaOH	350	ACS Catalysis 2013, 3 (2), 166-169
porous NiSe ₂ nanosheets	1 M KOH	184	Chem. Mater. 2015, 27 (16), 5702-5711
Ni ₅ P ₄ Films	1 M KOH	150	Angew. Chem., Int. Ed. 2015, 127(42), 12538-12542
Ni(OH) ₂ -Pt/C hybrid catalysts	1 M KOH	184	ACS Catalysis 2015, 5 (6), 3801-3806
NiFe LDHs	1 M KOH	219	Science 2014, 345 (6204), 1593-1596
CoP/CC	1 M KOH	209	J. Am. Chem. Soc. 2014, 136 (21), 7587-7590
Co NPs@N-CNTs	1 M KOH	370	Energy Environ. Sci. 2015, 8 (8), 2347-2351
Ni ₂ P	1 M KOH	220	Angew. Chem. Int. Ed., 2016, 55, 3804-3808
EG/Co _{0.85} Se/NiFeLDH	1 M KOH	300	Energy Environ Sci, 2016, 9, 478-483
NiCoFe LTHs/CFC	1 M KOH	200	ACS Energy Lett. 2016, 1, 445–453
NiCoFe-LTH/NF	1 M KOH	180	This work

Table S3. Comparison with other electrocatalysts for urea electrolysis reported in literatures

Electrocatalyst	Concentration of Urea (M)	Potential (mV) (10 mA cm ⁻²)	References
HC-NiMoS/Ti	0.5	1.66	Nano Res., 11 (2017) 988-996
Ni(OH) ₂ NS@NW/Ni foam nanosheets	0.33	1.58	Electrochim. Acta, 268 (2018) 211-217
Ni ₂ P/CFC	0.33	1.44	Electrochim. Acta, 254 (2017) 44-49
NiCo ₂ S ₄ NS/CC	0.33	1.45	ACS Sustain. Chem. Eng., 6 (2018) 5011-5020
Fe11.1%-Ni ₃ S ₂ /Ni foam	0.33	1.46	J. Mater. Chem. A, 6 (2018) 4346-4353
MnO ₂ /MnCo ₂ O ₄	0.5	1.58	J. Mater. Chem. A, 5 (2017) 7825-7832
CoS ₂ NA/Ti	0.30	1.59	Electrochim. Acta, 246 (2017) 776-782
Ni ₃ N NA/CC	0.33	1.44	Inorg. Chem. Front., 4 (2017) 1120-1124
NiFeCo LDH/NF	0.33	1.49	ACS Sustain. Chem. Eng., 7 (2019) 10035-10043
1% Cu:a-Ni(OH) ₂ /NF	0.33	1.49	J. Mater. Chem. A, 7 (2019) 13577-13584
NiCoFe-LTH/NF	0.33	1.49	This work