Supplementary Information

Facile surface engineering approach for fabricating the superwetting Ni-Fe-Co LDH@Ni-S heterojunction as bi-functional electrode for green hydrogen production: Experiment and theory

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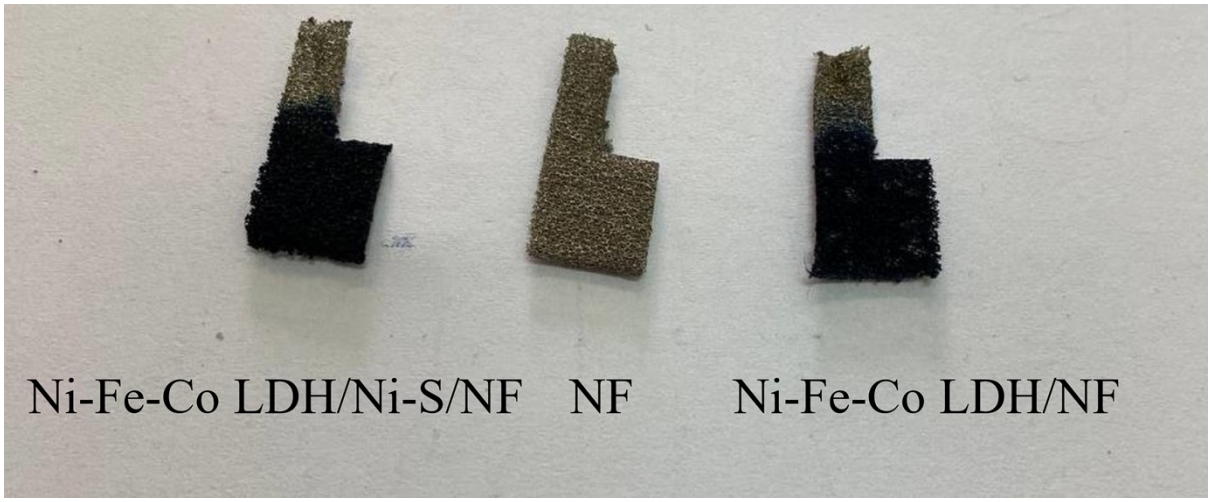


Figure S1. Pictures of Ni foam, Ni-Fe-Co/Ni-S/NF and Ni-Fe-Co/NF samples.

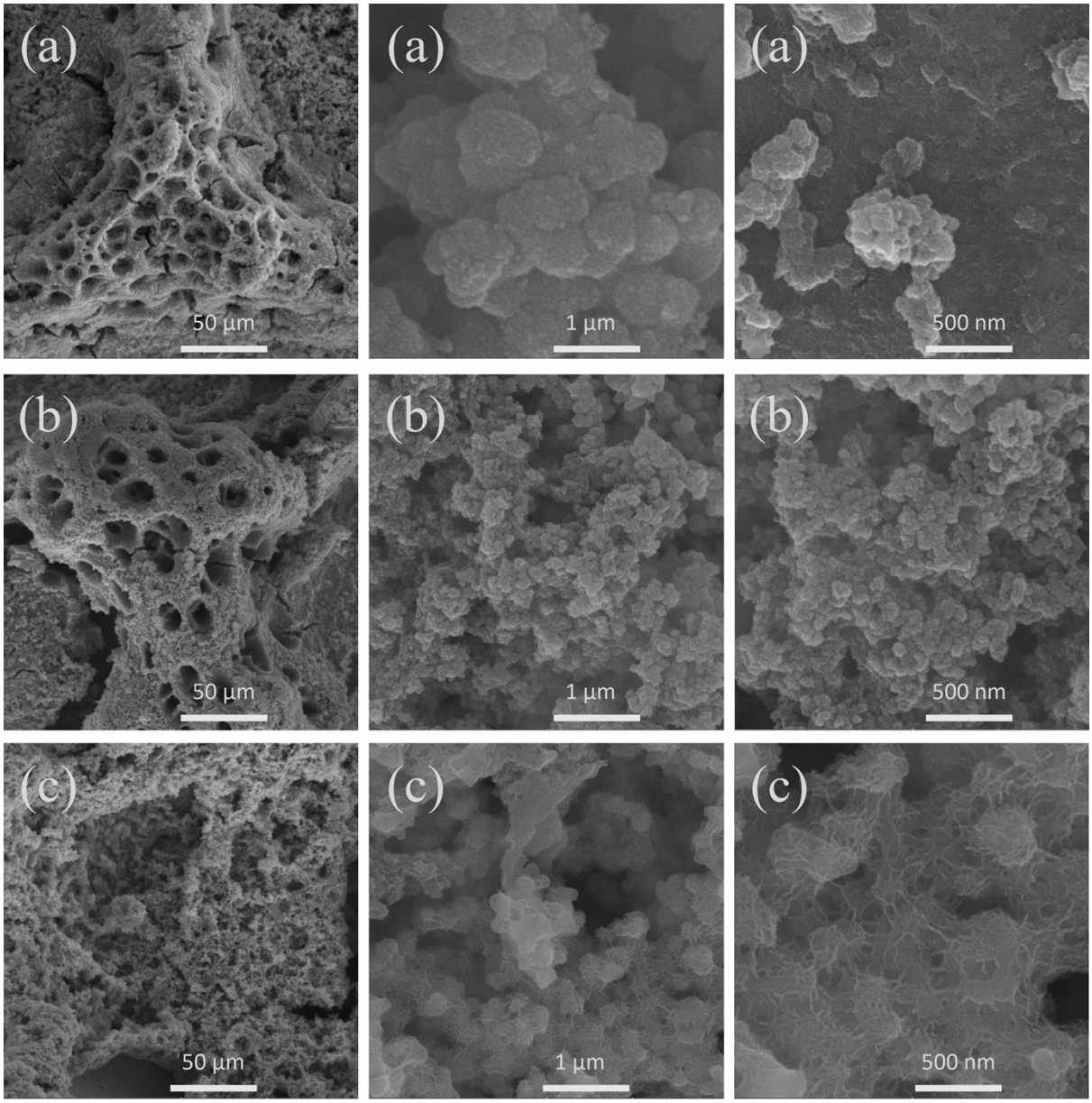


Figure S2. FESEM images of Ni-Fe-Co LDH/Ni-S/NF samples electrodeposited with (a) 1, (b) 3, (c) 10 cycles.

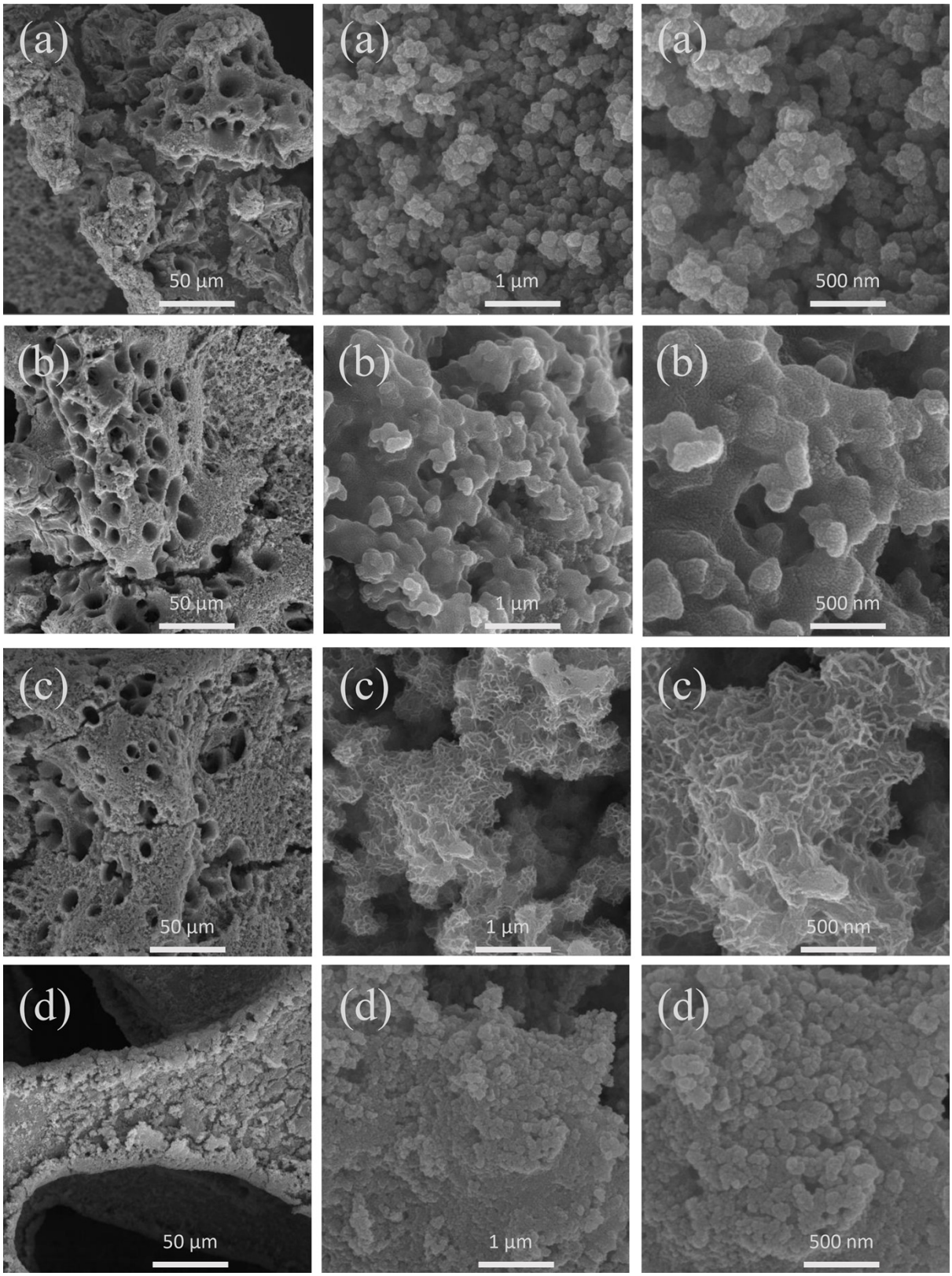
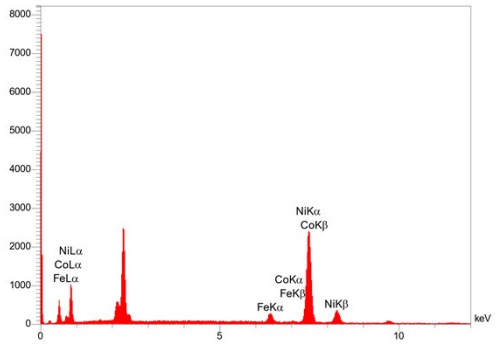


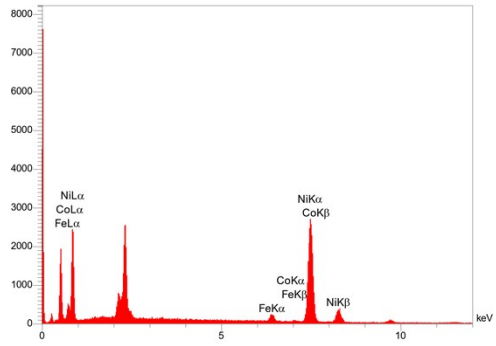
Figure S3. FESEM images of (a) Ni-Fe LDH/Ni-S/NF, (b) Ni-Co LDH/Ni-S/NF, (c) Fe-Co LDH/Ni-S/NF, (d) Ni-Fe-Co LDH/NF samples.

(a)



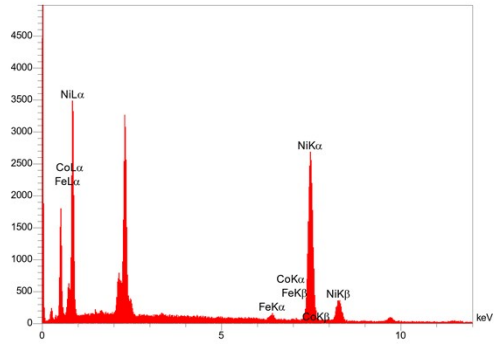
Element	Wt.%	At.%
Ni	93.70	93.45
Fe	5.47	5.73
Co	0.83	0.82

(b)



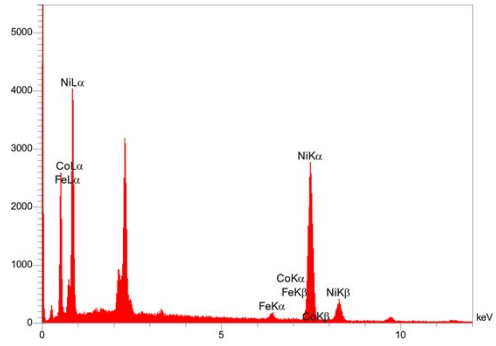
Element	Wt.%	At.%
Ni	94.13	93.96
Fe	3.63	3.81
Co	2.24	2.23

(c)



Element	Wt.%	At.%
Ni	94.50	94.46
Fe	1.20	1.26
Co	4.30	4.28

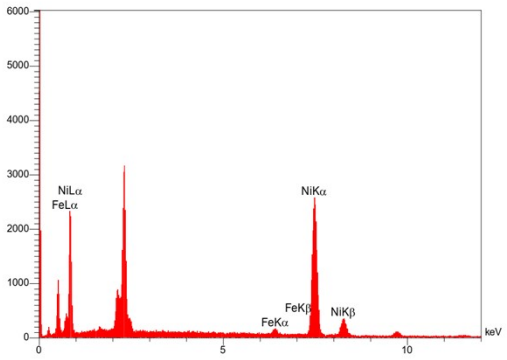
(d)



Element	Wt.%	At.%
Ni	93.13	93.07
Fe	1.71	1.79
Co	5.16	5.14

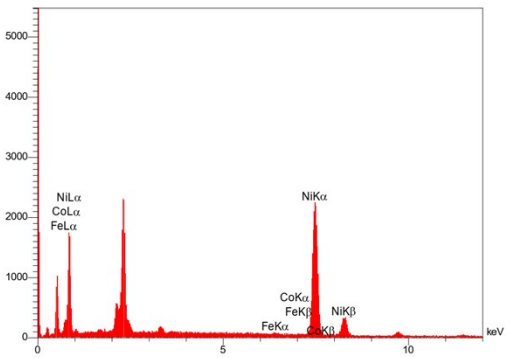
Figure S4. EDS results of Ni-Fe-Co LDH/Ni-S/NF samples electrodeposited with (a) 1, (b) 3, (c) 10 cycles.

(a)



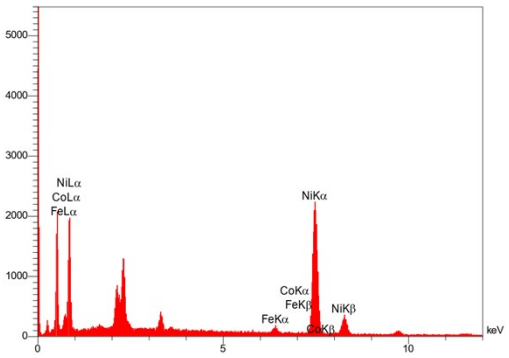
Element	Wt.%	At.%
Ni	98.21	98.12
Fe	1.79	1.88
Co	0	0

(b)



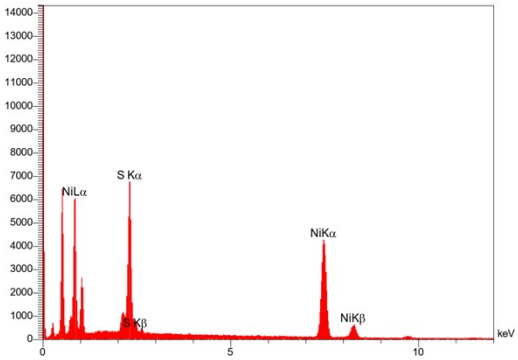
Element	Wt.%	At.%
Ni	97.63	97.63
Fe	0.17	0.18
Co	2.20	2.19

(c)



Element	Wt.%	At.%
Ni	94.69	94.60
Fe	2.02	2.13
Co	3.29	3.27

(d)



Element	Wt.%	At.%
Ni	49.83	35.17
S	50.17	64.83

Figure S5. EDS results of (a) Ni-Fe LDH/Ni-S/NF, (b) Ni-Co LDH/Ni-S/NF, (c) Fe-Co LDH/Ni-S/NF, (d) Ni-S/NF samples.

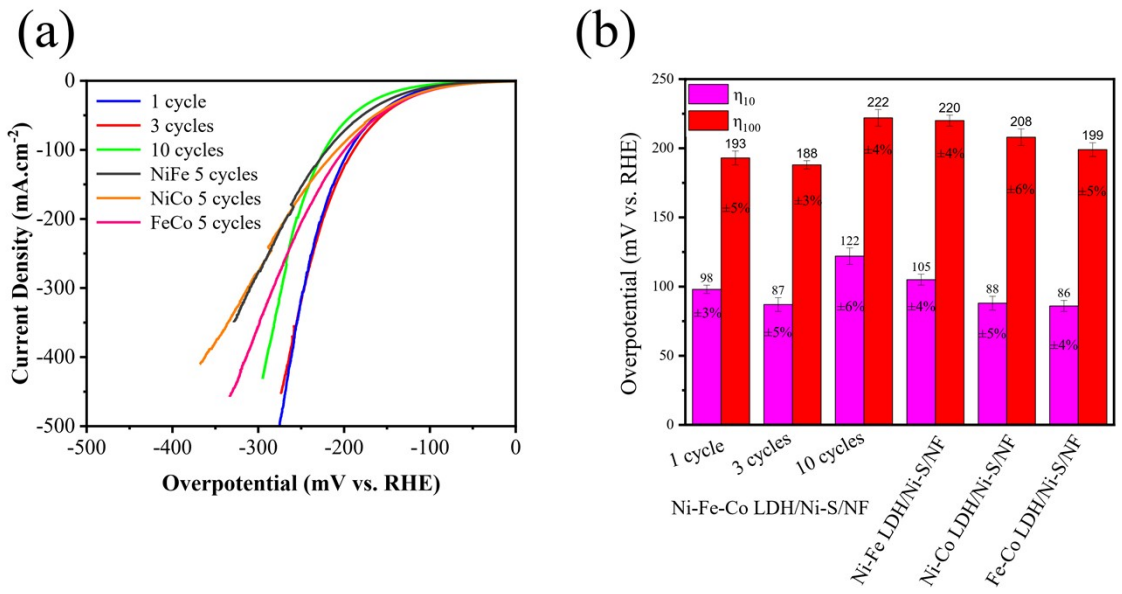


Figure S6. (a) LSV curves, (b) η_{10} and η_{100} values of Ni-Fe-Co/Ni-S/NF with different number of electrodeposition cycles and bi-elemental samples for HER in 1 M KOH + 0.5 M urea with scan rate of 5 mV s^{-1}.

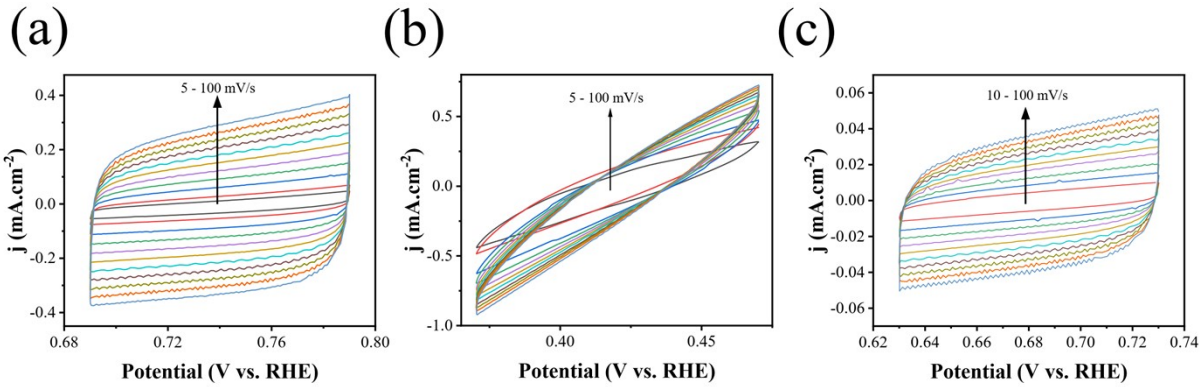


Figure S7. CV curves of (a) Ni-S/NF, (b) Ni-Fe-Co LDH/NF, (c) NF in non-faradic potential window of 0.1 V with different scan rates.

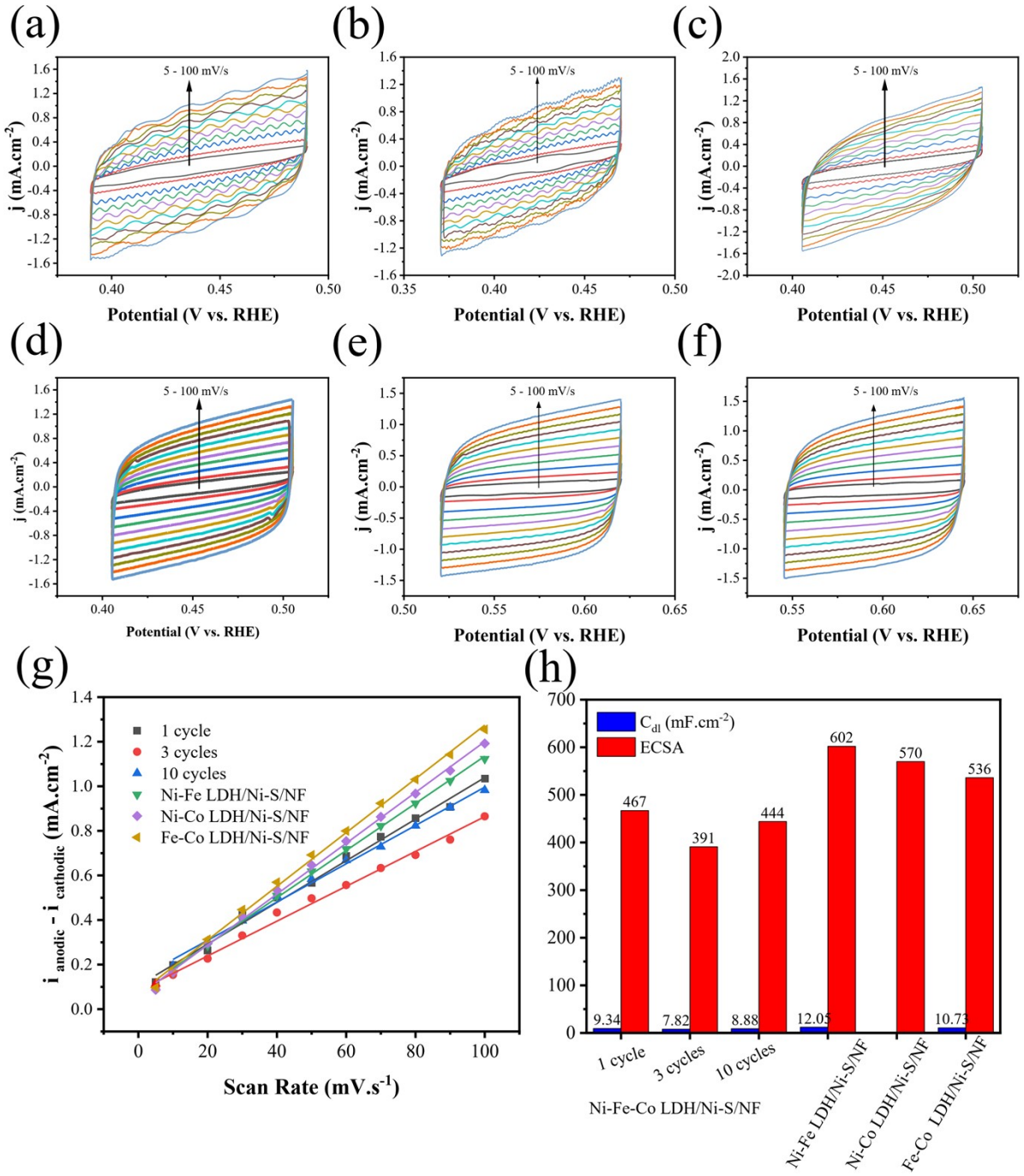
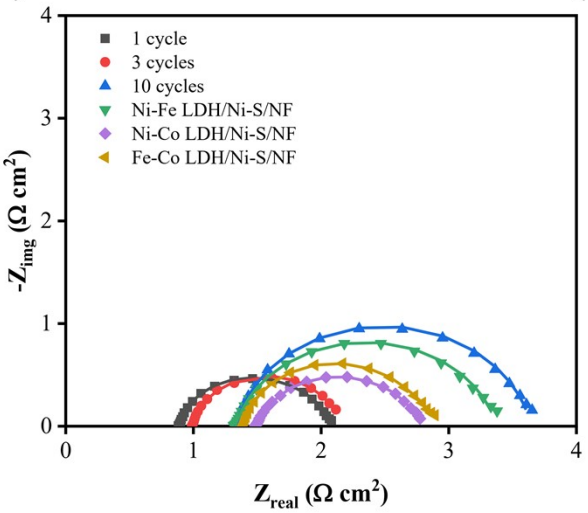


Figure S8. CV curves of Ni-Fe-Co LDH/Ni-S/NF samples electrodeposited with (a) 1, (b) 3, (c) 10 cycles, (d) Ni-Fe LDH/Ni-S/NF, (e) Ni-Co LDH/Ni-S/NF, (f) Fe-Co LDH/Ni-S/NF, (g, h) C_{dl} and ECSA values.

(a)



(b)

Catalyst	R_{ct} ($\Omega.cm^2$)
1 cycle	1.04
3 cycles	1.16
10 cycles	2.30
Ni-Fe LDH/Ni-S/NF	2.04
Ni-Co LDH/Ni-S/NF	1.30
Fe-Co LDH/Ni-S/NF	1.49

Figure S9. (a) Nyquist curves, (b) R_{ct} values of Ni-Fe-Co/Ni-S/NF with different number of electrodeposition cycles and bi-elemental samples for HER with overpotential of -200 mV vs. RHE.

(a)

(b)

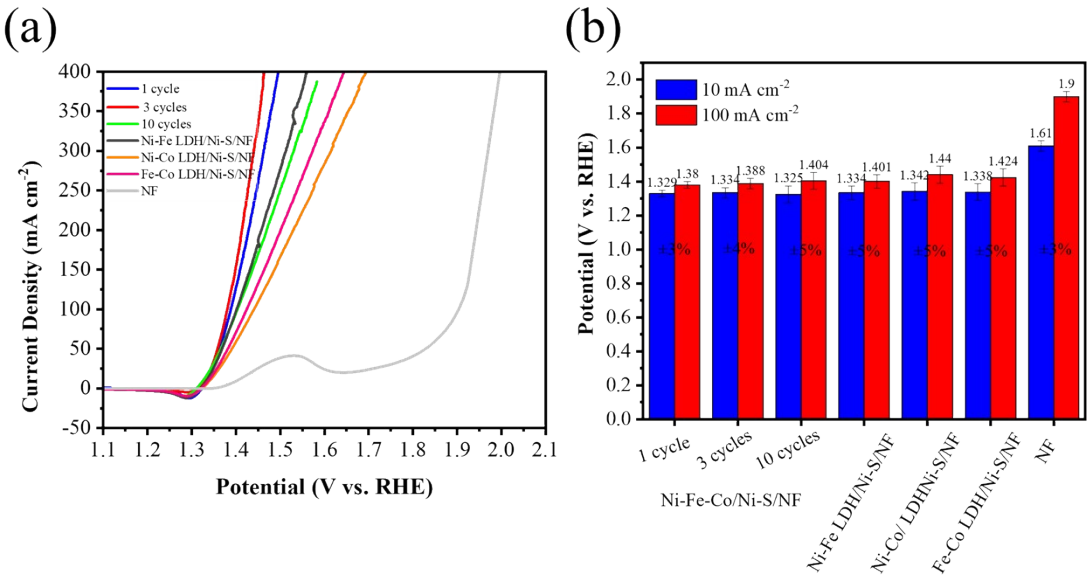
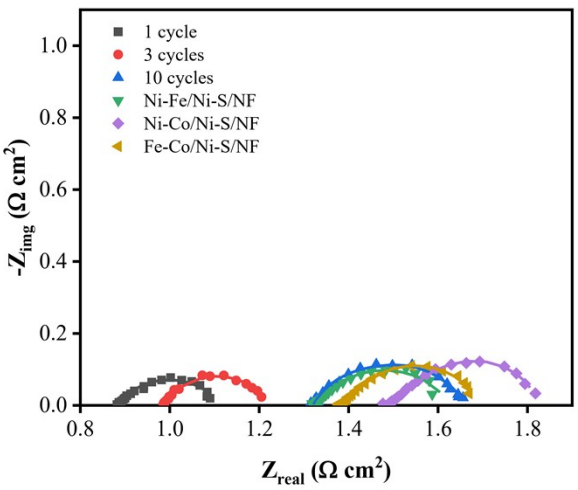


Figure S10. (a) LSV curves, (b) potential values in 10 and 100 mA cm^{-2} of Ni-Fe-Co/Ni-S/NF with different number of electrodeposition cycles and bi-elemental samples for HER in 1 M KOH + 0.5 M urea with scan rate of 5 mV s^{-1}.

(a)



(b)

Catalyst	R_{ct} ($\Omega.cm^2$)
1 cycle	0.22
3 cycles	0.23
10 cycles	0.31
Ni-Fe LDH/Ni-S/NF	0.29
Ni-Co LDH/Ni-S/NF	0.23
Fe-Co LDH/Ni-S/NF	0.25

Figure S11. (a) Nyquist curves, (b) R_{ct} values of Ni-Fe-Co/Ni-S/NF with different number of electrodeposition cycles and bi-elemental samples for UOR with potential of 1.4 V vs. RHE.

Table S1. EDS results of optimum Ni-Fe-Co/Ni-S/NF after CP.

Element	After HER		After UOR	
	Wt.%	At.%	Wt.%	At.%
Ni	95.04	95.05	95.57	95.53
Fe	1.10	1.11	1.10	1.11
Co	3.85	3.84	3.33	3.32

Table S2. Comparison of HER performance in different studies.

Catalyst	**Electrolyte**	**η_{10} for HER (mV vs. RHE)**	**Ref.**
This study	1 M KOH + 0.5 M urea	-73	-
NiFeCo LDH/NF	1 M KOH	-108	[1]
NiFeCoS	1 M KOH	-106	[2]
Ni-Fe-Co	1 M KOH	-91	[3]
Ni-Fe-Co graphene	6 M KOH	-169	[4]

(NiFe/NiCo)S@NF	1 M KOH	-185	[5]
Ni–Co–Fe–P	1 M KOH	-56	[6]
FeCoNiN/NF	1 M KOH	-56	[7]

Table S3. Comparison of UOR and OER performance in different studies.

Catalyst	**Electrolyte**	**Required potential in 10 mA cm^{-2} for UOR (V vs. RHE)**	**Required potential in 50 mA cm^{-2} for UOR (V vs. RHE)**	**Required potential in 100 mA cm^{-2} for UOR (V vs. RHE)**	**Cell voltage for 10 mA cm^{-2} (V)**	**Overpotential in 10 mA cm^{-2} for OER**	**Ref.**
This study	1 M KOH + 0.5 M urea	1.32	-	1.37	1.42	260	-
NiCo BMHs	1 M KOH + 0.33 M urea	1.33	-	-	-	-	[8]
Co-Ni_3S_2/FeS	1 M KOH + 0.33 M urea	-	-	-	1.50	-	[9]
FeCo LDH	1 M KOH	-	-	-	-	300	[10]
NiFeP/CoP	1 M KOH + 0.33 M urea & 1 M KOH	1.38	-	-	-	274	[11]
NiFeCo LDH/NF	1 M KOH + 0.33 M urea	1.34	-	-	1.49	210	[1]
$NiFeCoSe_2$	1 M KOH + 0.33 M urea	-	1.37	-	-	-	[12]
NiCoV LDH	1 M KOH + 0.33 M urea	1.33	-	-	1.45	-	[13]

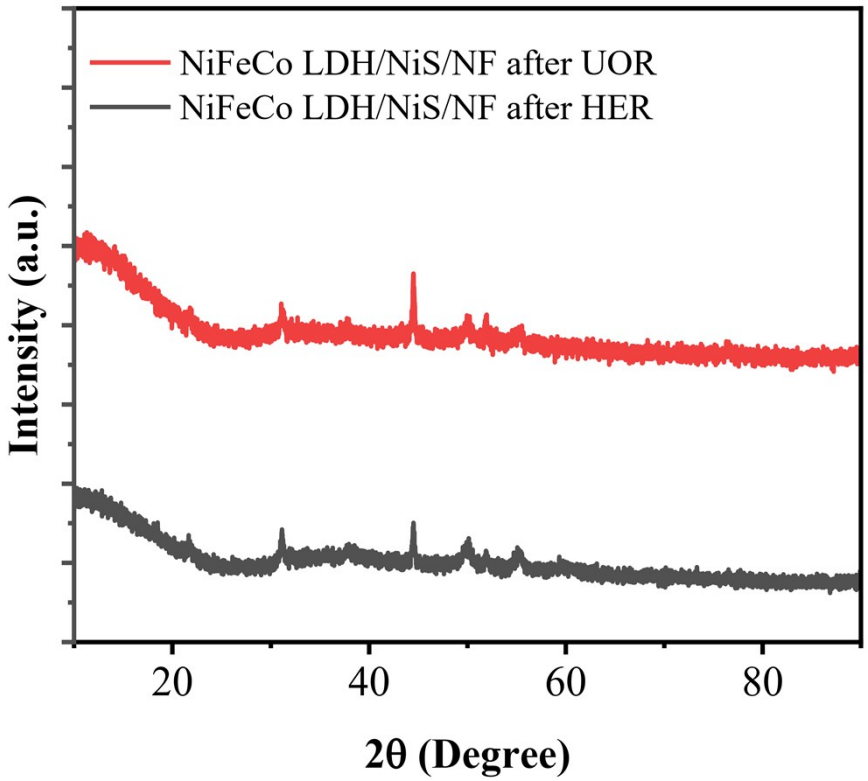


Figure S12. XRD pattern of Ni-Fe-Co LDH/Ni-S/NF after HER and UOR.

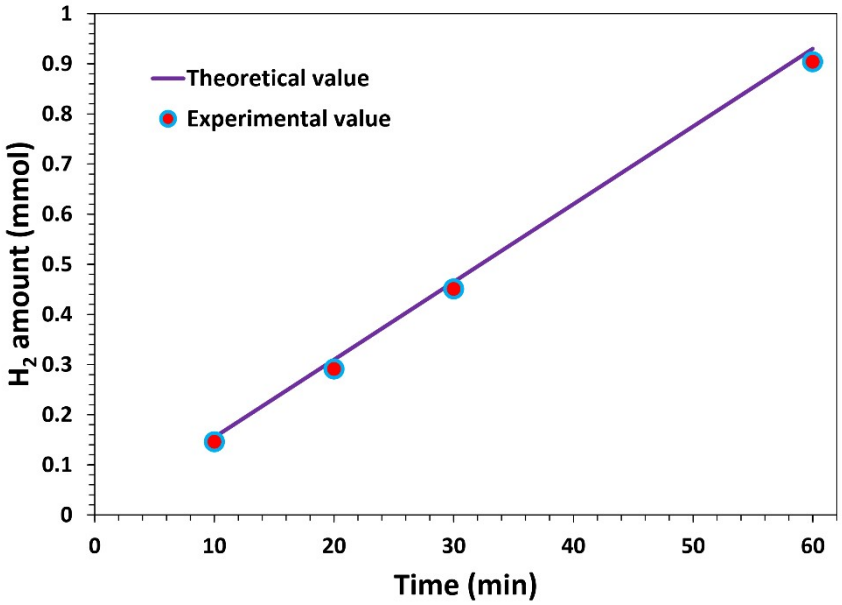


Figure S13. Calculated and measured hydrogen at 50 mA.cm^{-2} for obtaining of faradic efficiency.

References

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