Cauliflower-like NiFe alloys anchored on flake iron nickel carbonate hydroxide heterostructure towards superior overall water and urea electrolysis

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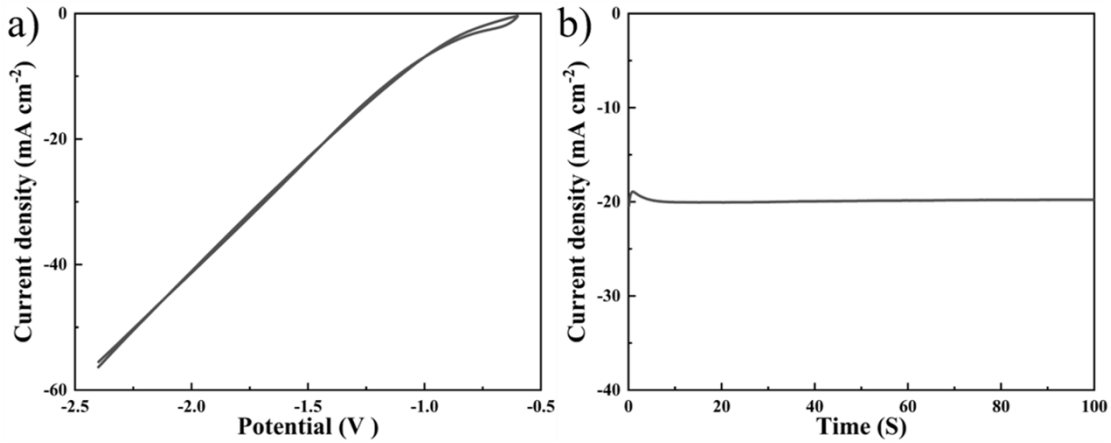

Fig.S1. The electrodeposition experiment of NiFe/NiFeCH/CC. (a) CV curve; (b) chronopotentiometric curve at a cathodic current density of 20 mA cm^{-2} for 100 s.

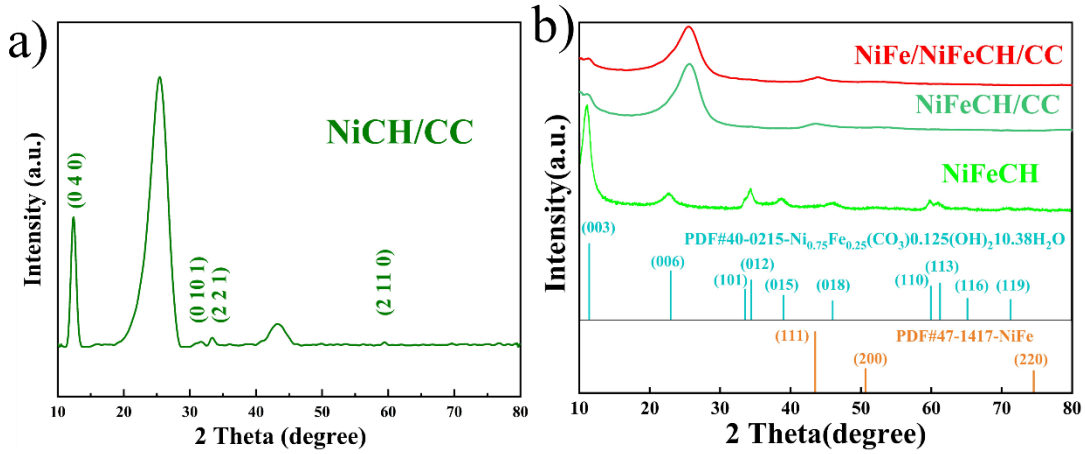

Fig.S2. (a) XRD pattern of NiCH/CC. (b) XRD patterns of powder NiFeCH, NiFeCH/CC and NiFe/NiFeCH/CC.

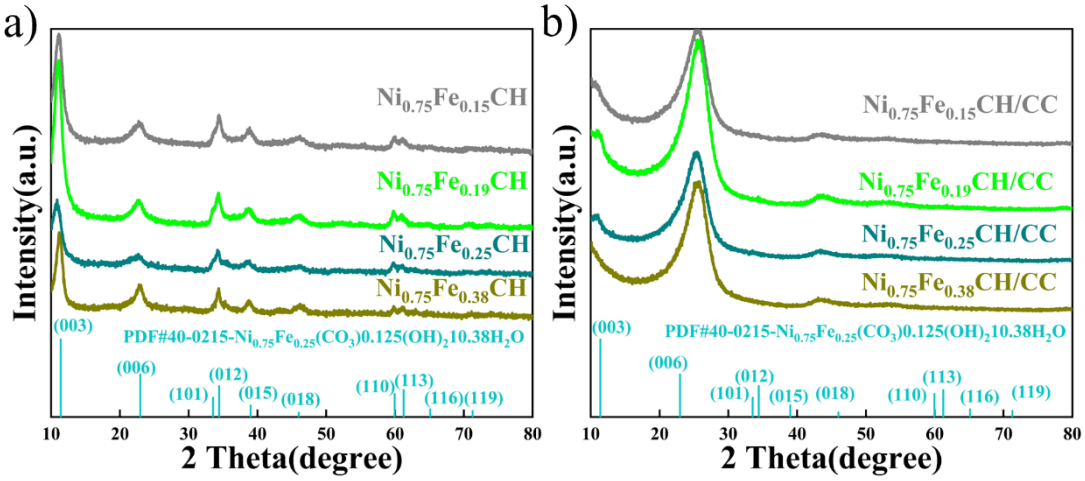

Fig.S3. (a) XRD patterns of powder NiFeCH collected from the precipitate after hydrothermal reaction with different proportions of Ni and Fe. (b) XRD patterns of the as-prepared NiFeCH/CC with different proportions of Ni and Fe.

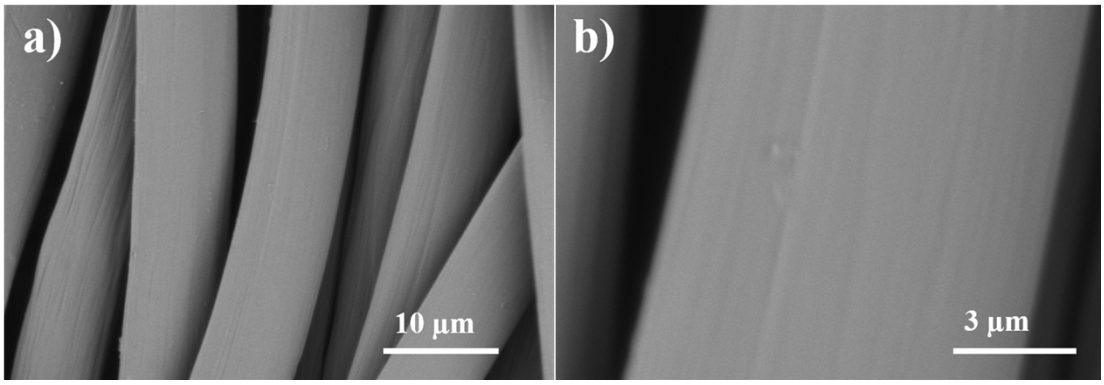

Fig.S4. SEM images of CC.

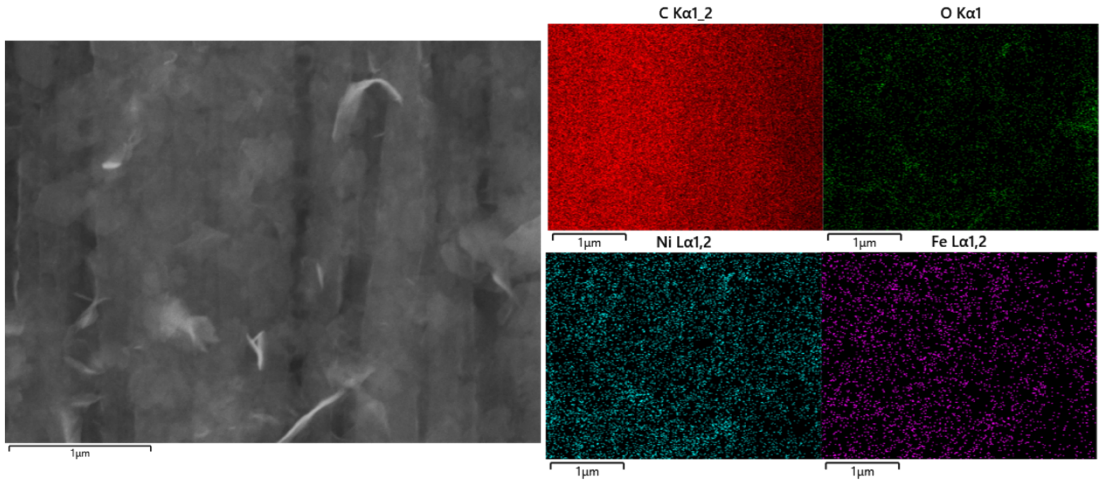

Fig.S5. Elemental mapping of NiFeCH/CC measured with SEM.

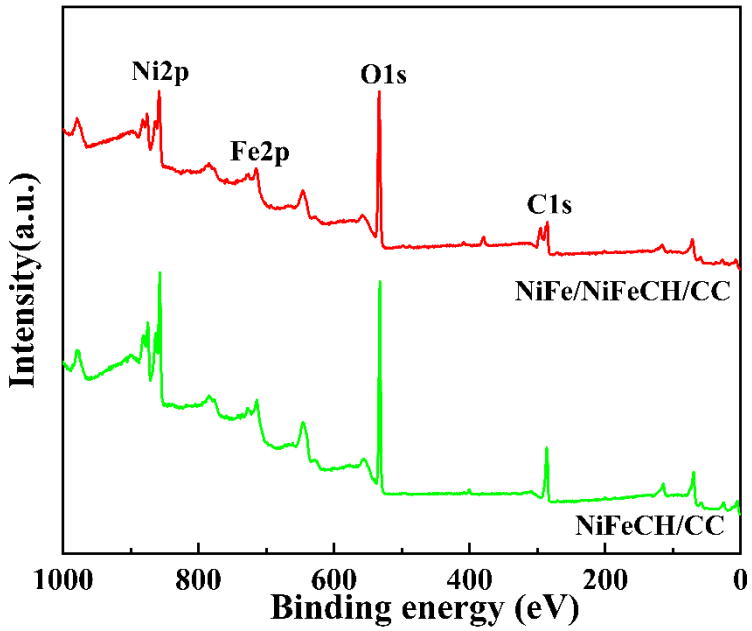

Fig.S6. XPS survey spectrum of NiFeCH and NiFe/NiFeCH/CC.

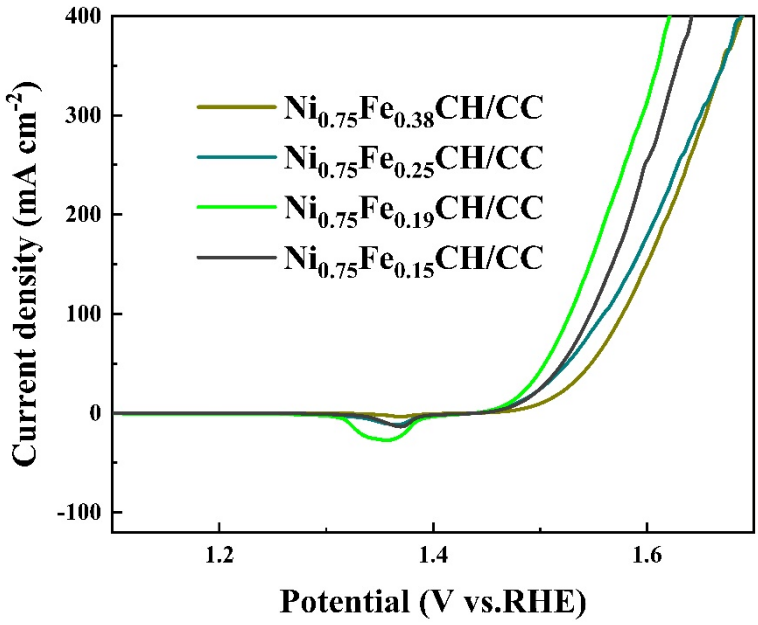

Fig.S7. LSV curves of NiFeCH/CC with different proportions of Ni and Fe.

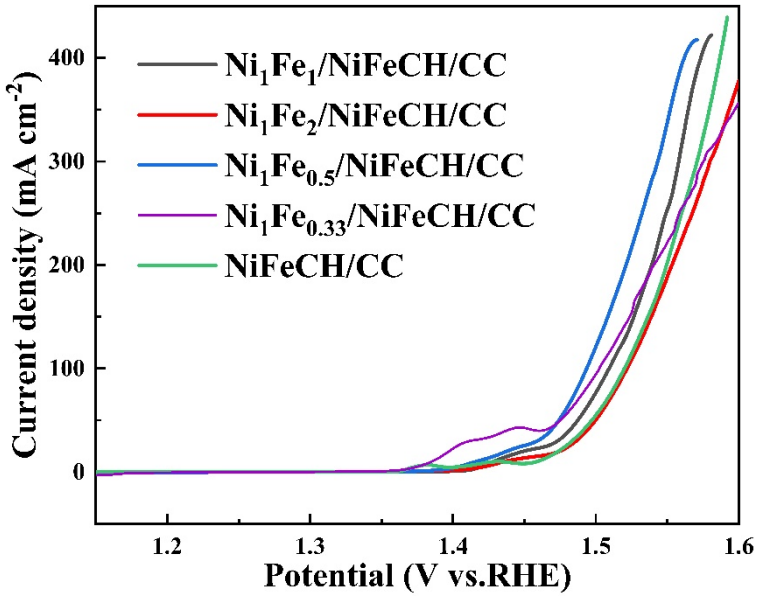

Fig.S8. LSV curves of NiFe/NiFeCH/CC with different proportions of Ni and Fe in NiFe alloy.

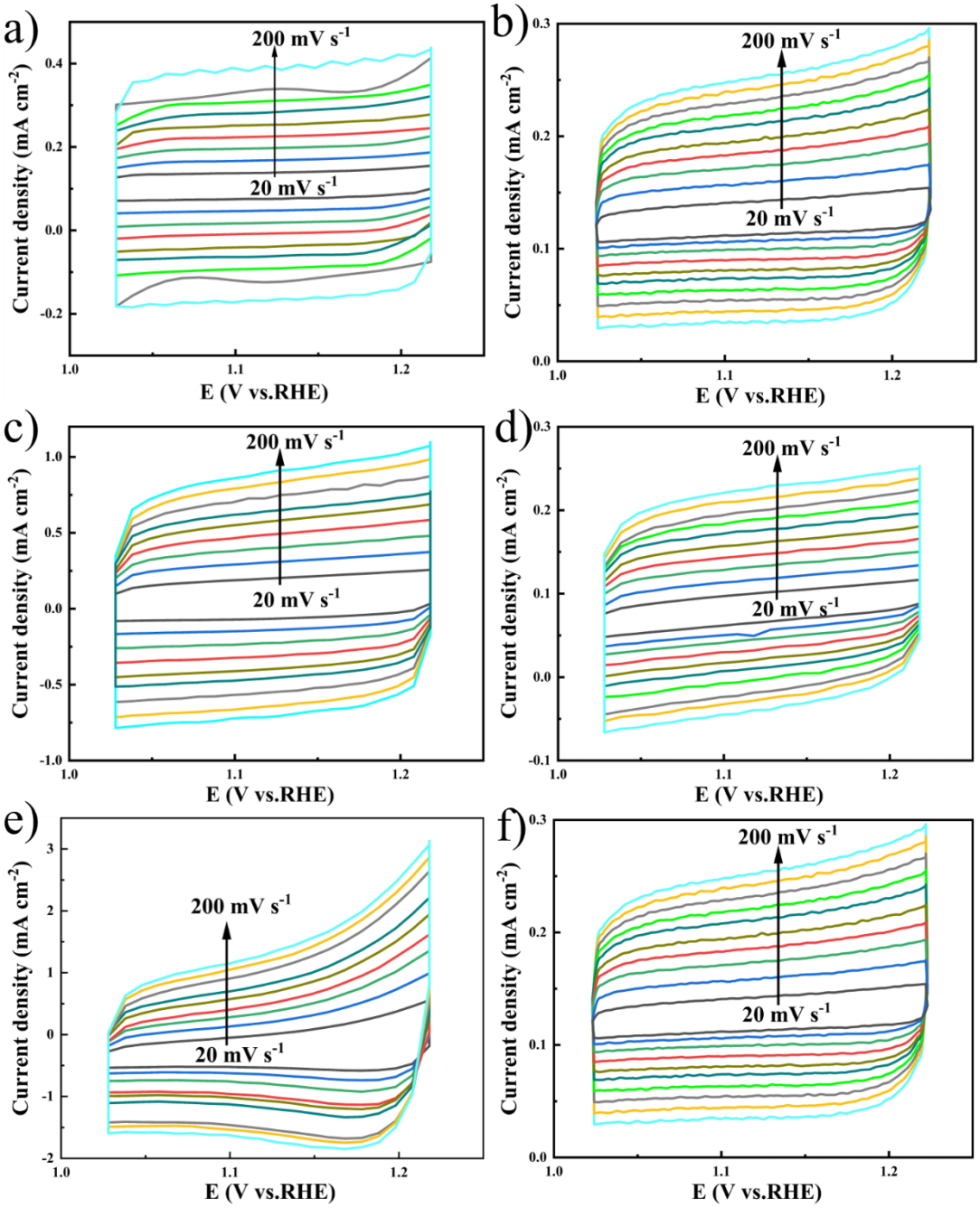

Fig.S9. CV curves with various scan rates. (a) NiCH/CC; (b) NiFeCH/CC; (c) Ni/NiFeCH/CC; (d) Fe/NiFeCH/CC; (e) NiFe/NiFeCH/CC; (f) NiFe/CC.

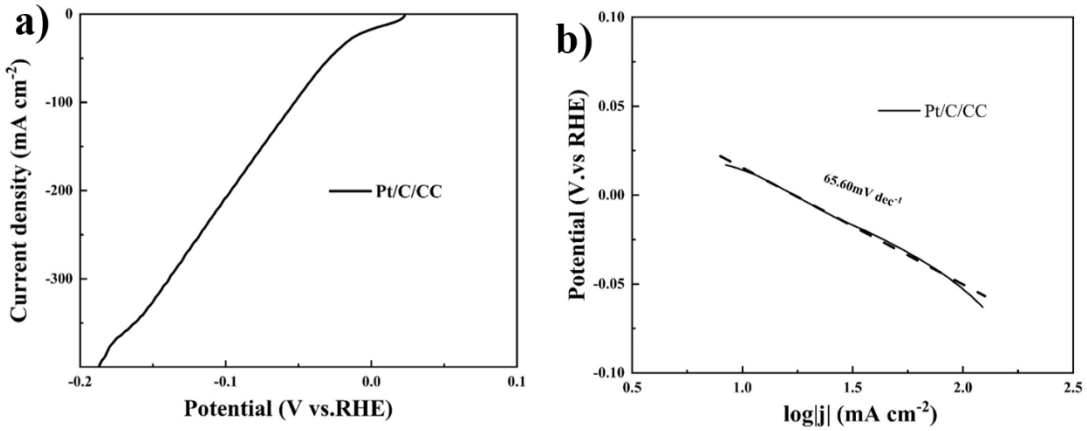

Fig.**S10**. (a) LSV curve and (b) corresponding Tafel plot of Pt/C/CC.

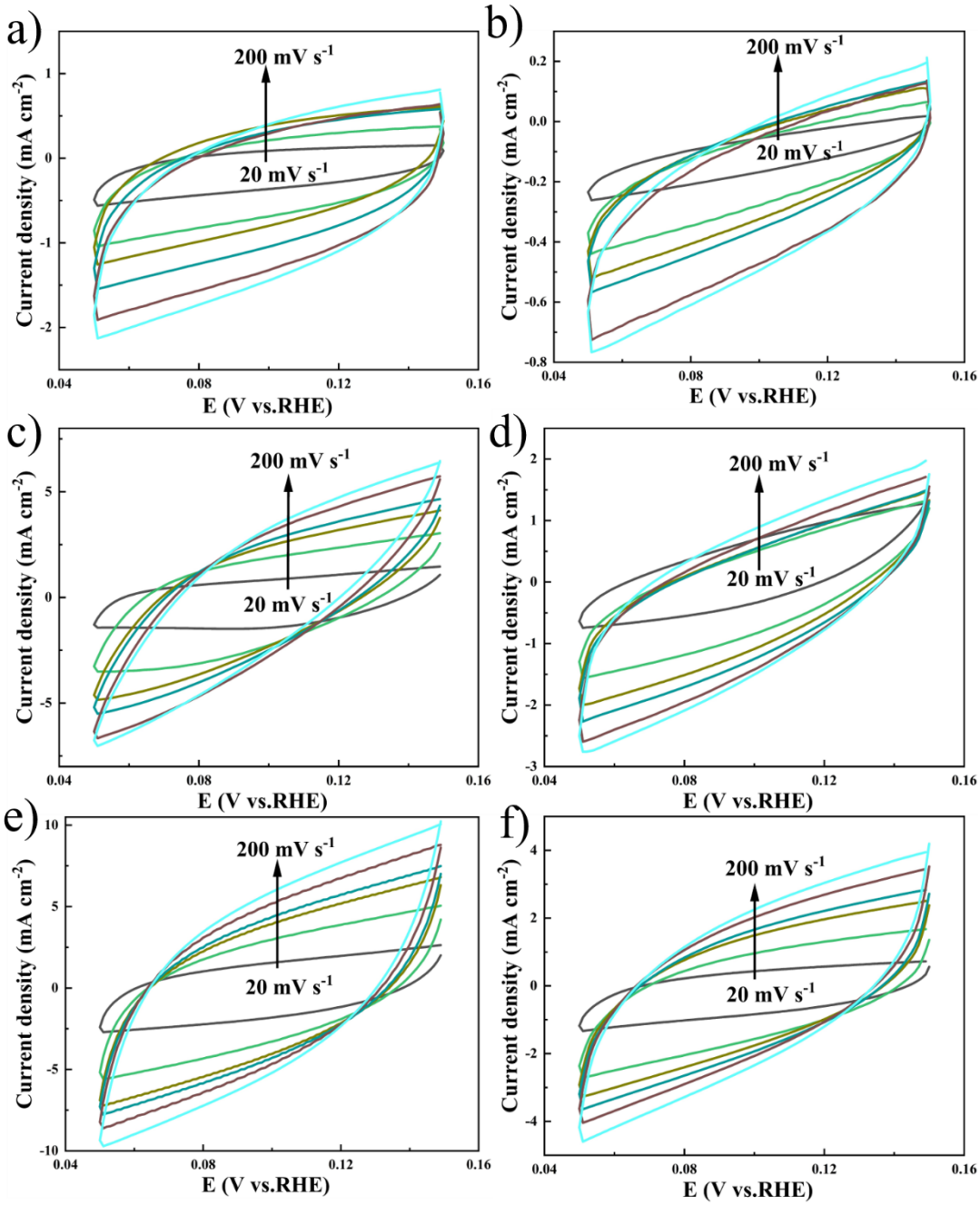

Fig.S11. CV curves with various scan rates. (a) NiCH/CC; (b) NiFeCH/CC; (c) Ni/NiFeCH/CC; (d) Fe/NiFeCH/CC; (e) NiFe/NiFeCH/CC; (f) NiFe/CC.

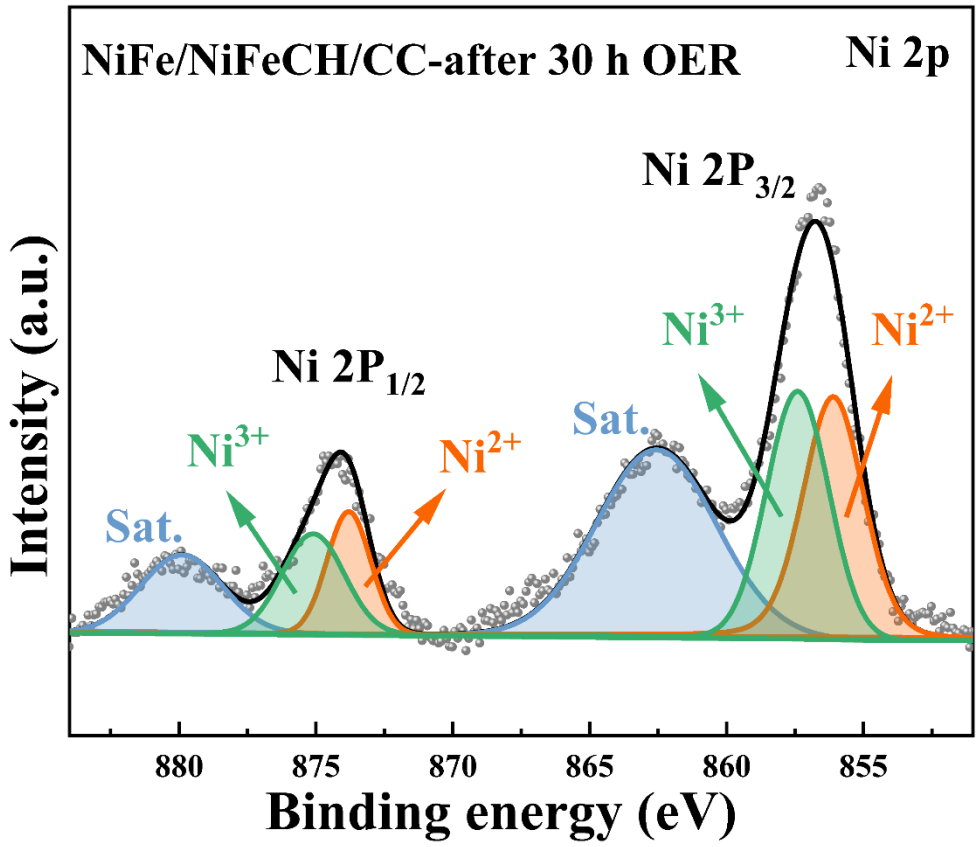

Fig.S12. High-resolution XPS spectra of Ni 2p before and after stability test.

Table S1. Content of $C_2H_3NiO_2 \cdot 4H_2O$ and $FeCl_3 \cdot 6H_2O$ added in hydrothermal reaction for as-prepared $Ni_{0.75}Fe_xCH$ electrocatalysts

Electrocatalysts	**$C_2H_3NiO_2 \cdot 4H_2O$ (mmol)**	**$FeCl_3 \cdot 6H_2O$ (mmol)**	**Ni: Fe**
$Ni_{0.75}Fe_{0.38}CH$	0.75	0.38	2:1
$Ni_{0.75}Fe_{0.25}CH$	0.75	0.25	3:1
$Ni_{0.75}Fe_{0.19}CH$	0.75	0.19	4:1
$Ni_{0.75}Fe_{0.15}CH$	0.75	0.15	5:1

Table S2. Content of $C_2H_3NiO_2 \cdot 4H_2O$ and $FeCl_3 \cdot 6H_2O$ added in electrochemical deposition reaction for as-prepared Ni_1Fe_x/NiFeCH electrocatalysts

Electrocatalysts	**$C_2H_3NiO_2 \cdot 4H_2O$ (mol/L)**	**$FeCl_3 \cdot 6H_2O$ (mol/L)**	**Ni: Fe**
Ni_1Fe_1/NiFeCH	0.01	0.01	1:1
Ni_1Fe_2/NiFeCH	0.01	0.02	1:2
$Ni_1Fe_{0.5}$/NiFeCH	0.01	0.005	2:1
$Ni_1Fe_{0.33}$/NiFeCH	0.01	0.0033	3:1

Table S3. ICP data for as-prepared $Ni_{0.75}Fe_x$CH electrocatalysts.

Electrocatalysts	**Ni (mmol)**	**Fe (mmol)**	**Ni: Fe (~)**
$Ni_{0.75}Fe_{0.38}$CH	$24.7*10^{-4}$	$8.25*10^{-4}$	3:1
$Ni_{0.75}Fe_{0.25}$CH	$23.2*10^{-4}$	$5.75*10^{-4}$	4:1
$Ni_{0.75}Fe_{0.19}$CH	$23.92*10^{-4}$	$4.6*10^{-4}$	5:1
$Ni_{0.75}Fe_{0.15}$CH	$24.3*10^{-4}$	$3.8*10^{-4}$	6:1

Table S4. Comparison of OER performance for NiFe/NiFeCH/CC with other bifunctional non-noble metal electrocatalysts tested in 1 M KOH.

Catalysts	Substrate	η (mV) 10 mA cm^{-2}	Stability (h)	Reference
NiFe/NiFeCH/CC	CC	210	150	This work
Ni_3FeN/r-GO	Ni foam	270	10	[1]
Co_9S_8@MoS_2/N-doped hollow carbon	/	233	12	[2]
Fe-NiCoP	Ni foam	235	18	[3]
CoFe/NF	Ni foam	220	50	[4]
NiFeOP	Ni foam	310	12	[5]
$Ni_{0.75}Fe_{0.25}Se_2$@NF	Ni foam	210	30	[6]
$CoFe_2O_4$/CoO-CNT	/	246	/	[7]
Co_2P/CoP@Co@NCNT	/	256	50	[8]
$CoSe_2$@$MoSe_2$	CC	183.8	/	[9]
Co_4Ni_1S/CC	CC	296	36	[10]
$Co(OH)_2$/NiMo CA@CC	CC	267	24	[11]
Co-Ni_3S_2	Ni foam	228	25	[12]
CoP@FeCoP/NC YSMPs	Carbon paper	238	20	[13]

Table S5. Comparison of HER performance for NiFe/NiFeCH/CC with other bifunctional non-noble metal electrocatalysts tested in 1 M KOH.

Catalysts	Substrate	η (mV) 10 mA cm^{-2}	Stability (h)	Reference
NiFe/NiFeCH/CC	CC	90	30	This work
Ni_3FeN/r-GO	Ni foam	94	10	[1]
Co_9S_8@MoS_2/N-doped hollow carbon	/	126	12	[2]
Fe-NiCoP	Ni foam	147	20	[3]
CoFe/NF	Ni foam	110	50	[4]
NiFeOP	Ni foam	209	14	[5]
$Ni_{0.75}Fe_{0.25}Se_2$@NF	Ni foam	117	30	[6]
$CoFe_2O_4$/CoO-CNT	/	164	/	[7]
Co_2P/CoP@Co@NCNT	/	118	12	[8]
$CoSe_2$@$MoSe_2$	CC	109.9	/	[9]
Co_4Ni_1S/CC	CC	192	32	[10]
$Co(OH)_2$/NiMo CA@CC	CC	30	24	[11]
Co-Ni_3S_2	Ni foam	102	25	[12]
CoP@FeCoP/NC YSMPs	Carbon paper	141	20	[13]

Table S6. Comparison of overall water splitting performance for NiFe/NiFeCH/CC with other bifunctional non-noble metal electrocatalysts tested in 1 M KOH.

Catalysts	Substrate	E_{10} (V)	Stability (h)	Reference
NiFe/NiFeCH/CC	CC	1.49	30	This work
Ni_3FeN/r-GO	Ni foam	1.60	100	[1]
Co_9S_8@MoS_2/N-doped hollow carbon	/	1.56	12	[2]
Fe-NiCoP	Ni foam	1.60	18	[3]
CoFe/NF	Ni foam	1.64	50	[4]
NiFeOP	Ni foam	1.69	20	[5]
$Ni_{0.75}Fe_{0.25}Se_2$@NF	Ni foam	1.61	50	[6]
$CoFe_2O_4$/CoO-CNT	/	1.61	~60	[7]
Co_2P/CoP@Co@NCNT	/	1.60	12	[8]
$CoSe_2$@$MoSe_2$	CC	1.53	24	[9]
Co_4Ni_1S/CC	CC	1.60	~10	[10]
$Co(OH)_2$/NiMo CA@CC	CC	1.52	30	[11]
Co-Ni_3S_2	Ni foam	1.54	25	[12]
CoP@FeCoP/NC YSMPs	Carbon paper	1.68	20	[13]

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