Electronic Supporting information

Highly Active Nickel-Cobalt/Nanocarbon Thin Films as Efficient Water Splitting Electrodes

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Fig. S1 (a) Optical image of the Pulsed laser deposition instrument and (b) Five designed bimetallic targets to induce deposition of deferent compositions of Ni_xCo_{1-x} species.



Fig. S2 XRD spectra of Ni/NC, Ni_xCo_{1-x}/NC and Co/NC.



Fig. S3 SEM top-view of N-carbon and Ni_xCo_{1-x}/NC films.



Fig. S4 (a) TEM image (inset of a) HR-TEM image and (b) TEM-EDS spectrum of Ni_xCo_{1-x}/NC film.



Fig. S5 High-resolution N1s spectrum of Ni_xCo_{1-x}/NC film.



Fig. S6 Deconvoluted peaks of the D-G region in the Raman spectra of (a) Ni/NC, (b) $Ni_{0.5}Co_{0.5}/NC$ and (c) Co/NC.



Fig. S7 Raman spectra (metal oxide region) of Ni/NC, Ni_{0.5}Co_{0.5}/NC and Co/NC.



Fig. S8 HER Tafel plots of $Ni_{0.7}Co_{0.3}/NC$, $Ni_{0.5}Co_{0.5}/NC$, $N_{0.4}Co_{0.6}/NC$, $Ni_{0.3}Co_{0.7}/NC$ and $Ni_{0.2}Co_{0.8}/NC$ in 1.0 M KOH solution.



Fig. S9 Nyquist plots of binary Ni_xCo_{1-x}/NC at a potential of -1.5 V (vs. Ag/AgCl).



Fig. S10 Chronoamperometric response of Ni _{0.5}Co_{0.5}/NC under a constant voltage of -1.5 V (vs. Ag/AgCl) at 1.0M KOH (inset) enlarged view of the working electrode during electrochemical tests.



Fig. S11 OER Tafel plots of $Ni_{0.7}Co_{0.3}/NC$, $Ni_{0.5}Co_{0.5}/NC$, $N_{0.4}Co_{0.6}/NC$, $Ni_{0.3}Co_{0.7}/NC$ and $Ni_{0.2}Co_{0.8}/NC$ in 1.0 M KOH solution.



Fig. S12 Chronoamperometric response of $Ni_{0.5}Co_{0.5}/NC$ under a constant voltage of +0.8 V (vs Ag/AgCl) in 1.0M KOH (inset) enlarged view of the working electrode during electrochemical tests.



Fig. S13 (a) Full range OER and HER LSV of $Ni_{0.5}Co_{0.5}/NC$ and (b) comparison of overall water splitting ability of the synthesized electrocatalysts.



Fig. S14 Cyclic voltammograms (CVs) for (a) Ni/NC, (b) $Ni_{0.5}Co_{0.5}/NC$, (c) carbon-free $Ni_{0.5}Co_{0.5}$ and (d) Co/NC measured at different scan rates from 10 to 100 mV s⁻¹ and (e) corresponding plots of the current density at -0.775 V vs. the scan rate.



Fig. S15 Cyclic voltammograms (CVs) for (a) $Ni_{0.7}Co_{0.3}/NC$, (b) $Ni_{0.4}Co_{0.6}/NC$, (c) $Ni_{0.3}Co_{0.7}/NC$ and (d) $Ni_{0.2}Co_{0.8}/NC$ measured at different scan rates from 10 to 100 mV s⁻¹ and (e) corresponding plots of the current density at -0.775 V vs. the scan rate.

Sample	С	0	Ν	Ni	Со	% Ni	% Co
Ni/NC	52.56	34.12	3.67	9.65	0	100	0
Ni _{0.7} Co _{0.3} /NC	54.72	33.49	2.77	6.18	2.84	69	31
Ni _{0.5} Co _{0.5} /NC	49.63	35.58	3.36	5.47	5.96	48	52
Ni _{0.4} Co _{0.6} /NC	49.33	36.31	3.04	4.14	7.18	38	62
Ni _{0.3} Co _{0.7} /NC	49.85	35.02	2.4	4.13	8.60	32	68
Ni _{0.2} Co _{0.8} /NC	48.09	37.76	1.9	9.61	2.64	78	22
Co/NC	52.07	34.33	3.91	0	9.69	0	100

Table S1 Chemical composition details obtained from XPS analysis for all synthesized

 electrodes

Table S2 Peak positions of Raman spectra for Ni/NC, Ni_0.5Co_0.5/NC and Co/NC.

Sample	D band position (cm ⁻¹)	G band position (cm ⁻¹)	I _D /I _G
Ni/NC	1373	1543	0.52
Ni _{0.5} Co _{0.5} /NC	1376	1551	0.61
Co/NC	1377	1549	0.58

Samples	ηο (V)	η10 (V)	Tafel Slope	Reference
Ni/NC	0.184	0.406	218.8	This work
Ni _{0.7} Co _{0.3} /NC	0.110	0.303	148.7	This work
Ni _{0.5} Co _{0.5} /NC	0.048	0.176	132.1	This work
Ni _{0.4} Co _{0.6} /NC	0.088	0.225	163.6	This work
Ni _{0.3} Co _{0.7} /NC	0.116	0.276	165.8	This work
Ni _{0.2} Co _{0.8} /NC	0.161	0.367	193.2	This work
Co/NC	0.163	0.378	221.9	This work
Ni0.5Co0.5	0.077	0.282	189.3	This work
Ni ₃ S ₂ /MWCNTs	>0.200	0.480	167	Ref 1
Co@NRCNT	160	370	80	Ref 2
NiO/Ni-CNT	90	~100	82	Ref 3
Co@N-C	125	200	100	Ref 4
Ni ₂ P	95	230	87	Ref 5

 Table S3 Summary of recently reported HER electrocatalysts in 1.0 M KOH.

Samples	η0 (V)	η10 (V)	Tafel Slope	Reference
Ni/NC	1.547	1.650	100.0	This work
Ni _{0.7} Co _{0.3} /NC	1.470	1.567	76.3	This work
Ni _{0.5} Co _{0.5} /NC	1.468	1.530	62.9	This work
Ni _{0.4} Co _{0.6} /NC	1.472	1.558	80.4	This work
Ni _{0.3} Co _{0.7} /NC	1.473	1.572	80.9	This work
Ni _{0.2} Co _{0.8} /NC	1.477	1.579	82.6	This work
Co/NC	1.524	1.614	136.2	This work
Ni _{0.5} Co _{0.5}	1.502	1.623	91.6	This work
Co _{1-x} Fe _x S@N-MC	1.570	1.640	159	Ref 6
N-graphene-NiCo ₂ O ₄	1.540	1.664	156.0	Ref 7
NiCo ₂ O ₄ NNs on FTO	1.595	1.795	292.0	Ref 8
NiCo ₂ O ₄ nanowire	1.520	1.550	63.1	Ref 9
Ni-Co ₂ -O	>1.500	1.592	64.4	Ref 10
Co ₃ O ₄ -NrmGO	1.509	1.540	67.0	Ref 11
N-CG-CoO	1.514	1.570	71.0	Ref 12
Co ₃ O ₄ /MWCNT	1.585	1.840	65.0	Ref 13

Table S4 Summary of recently reported OER electrocatalysts in 1.0 M KOH.

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