Electronic Supplementary Information

## Simultaneous phase transformation and doping via a unique photochemical-electrochemical strategy to highly active Fedoped Ni oxyhydroxide oxygen evolution catalyst

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Fig. S1 Schematic depiction of the synthesis of *pc*-NiFe@CC electrocatalyst.



Fig. S2 CV scans of *a*-NiO<sub>x</sub>@CC in 5  $\mu$ M Fe<sup>3+</sup> spiked 1.0 M KOH, scan rate: 100 mV s<sup>-1</sup>.



Fig. S3 SEM images of bare CC. a, low magnification; b, high magnification.



Fig. S4 SEM images of Ni-PDA@CC. a, high magnification; b, low magnification.



Fig. S5 SEM images of *a*-NiO<sub>x</sub>@CC. a, high magnification; b, low magnification.



Fig. S6 TEM image of nanosheets ultrasonically peeled from Fe-NiOOH@CC.



**Fig. S7** TEM images of ultrasonically peeled a-NiO<sub>x</sub>. a, low magnification; b, high magnification.



**Fig. S8** OER activity of Fe-NiOOH@CC synthesized with different duration of Ni-PDA deposition.



Fig. S9 OER activity of Fe-NiOOH@CC synthesized with different duration of UV irradiation.



**Fig. S10** OER activity of Fe-NiOOH@CC synthesized with different Fe spiking concentration for cycling.



**Fig. S11** CV curves of Fe-NiOOH@CC (a), *ec*-NiFe@CC (b), *pc*-NiFe@CC (c) and NiOOH@CC (d) in 1.0 M KOH solution at different scanning rates.



Fig. S12 ECSA-normalized LSV curves of Fe-NiOOH@CC, ec-NiFe@CC, pc-NiFe@CC and NiOOH@CC with iR compensation. The ECSA of catalysts is

calculated from the double-layer capacitance according to equation:  $ECSA = \frac{C_{dl}}{C_s},$ 

where  $C_{dl}$  is the double-layer capacitance, estimated by the slope of a linear curve that obtained by plotting the non-Faradaic capacitive current ( $\Delta J/2$ , i.e. ( $J_a - J_c$ )/2) against scanning rate;  $C_s$  is the specific capacitance of a planar surface. The  $C_s$  value is selected as 0.04 mF cm<sup>-2</sup> in present work (see e.g., J. Am. Chem. Soc., 2013, 135, 16977).



Fig. S13 TEM images of Fe-NiOOH@CC after OER electrolysis.



Fig. S14 Ni 2p (a) and Fe 2p (b) XPS spectra of Fe-NiOOH@CC after OER.



Fig. S15 XPS Fe 2p spectra of NiOOH@CC, pc-NiFe@CC and ec-NiFe@CC.



Fig. S16 XRD patterns of NiOOH@CC, pc-NiFe@CC and ec-NiFe@CC.



**Fig. S17** LSV curves of Fe-NiOOH@ITO and Fe-NiOOH@FTO at 5 mV s<sup>-1</sup> in 1.0 M KOH.

Catalyst	η@10 mA cm <sup>-2</sup> (mV)	Tafel slope (mV dec <sup>-1</sup> )	Stability	Electrolyte	Morphology	Substrate	Ref.
Fe-NiOOH@CC	331	64	150 mA cm <sup>-2</sup> for 55 h	1.0 M KOH	nanoparticle	CC	This work
r-FeOOH/α-Ni(OH) <sub>2</sub> /NF	168	51.4	2000 mA cm <sup>-2</sup> for 10 h	1.0 M KOH	particle	NF <sup>a</sup>	1
NiFe LDH@NiCoP/NF	220	48.6	10 mA cm <sup>-2</sup> for 100 h	1.0 M KOH	nanosheet on nanowire	NF	2
NiFe LDH/NiTe	228 @50 mA cm <sup>-2</sup>	51.04	20 mA cm <sup>-2</sup> for 30 h	1.0 M KOH	nanosheet on nanorod	NF	3
Ni-Fe-OH@Ni <sub>3</sub> S <sub>2</sub> /NF	165	93	500 mA cm <sup>-2</sup> for 50 h	1.0 M KOH	nanosheet	NF	4
NiFe/N-CNT	290	79	5000 cycles	0.1 M KOH (1600 rpm)	nanoparticle	CNT <sup>b</sup>	5

**Table S1**. OER activity of NiFe-base catalysts in alkaline media reported in recent literature.

CoFe@NiFe-200/NF	190	45.71	36 mA cm <sup>-2</sup> for 30 h	1.0 M KOH	nanosheet on nanosheet	NF	6
NiFe-LDH/NF	270	67.44	/	1.0 M KOH	nanosheet	NF	6
NiFe LDH	182	34.34	1.7 V for 100 h	1.0 M KOH	nanosheet	NF	7
NiFeRu-LDH	225	32.4	10 mA cm <sup>-2</sup> for 10 h	1.0 M KOH	nanosheet	NF	8
NiFe-LDH	230	36.2	N.A.	1.0 M KOH	nanosheet	NF	8
MIL-53(FeNi)/NF	233 @50 mA cm <sup>-2</sup>	31.3	20, 50, 100 mA cm <sup>-2</sup> for 16000 s, respectively	1.0 M KOH	nanosheet	NF	9
4.3%-strained NiFe MOFs	210 @200 mA cm <sup>-2</sup>	68	200 mA cm <sup>-2</sup> for 200 h	0.1 M KOH	nanosheet	NF	10
pristine NiFe MOFs	600 @200 mA cm <sup>-2</sup>	167	N.A.	0.1 M KOH	nanosheet	NF	10

a-LaNiFe(t-d) hydroxide	189	36	10 mA cm <sup>-2</sup> for 100 h	1.0 M KOH	nanostructured amorphous	NF	11
ball-milled NiFe-LDH	270	36.2	N.A.	1.0 M KOH	nanosheet	GC °	12
NiFe-NFF	227	38.9	10~20 mA cm <sup>-2</sup> for 15 h	1.0 M KOH	nanocluster on nanosheet	NFF <sup>d</sup>	13

<sup>a</sup> NF: nickel foam, <sup>b</sup> CNT: carbon nanotubes, <sup>c</sup> GC: glassy carbon electrode, <sup>d</sup> NFF: NiFe alloy foam.

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