Etch-Induced Ion Exchange Engineering of Two-Dimensional Layered NiFeCo-Based Hydroxides for High-Energy Charge Storage

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Figure Captions and Video

- Fig. S1 Photographic images of the beginning and ending of the electrochemical synthesis process, and of pure NF, NF after etching treatment, and NF after electrochemical synthesis.
- Fig. S2 Photographic images of pre-treatment pure NF (a), Ni/NF (b), Ni₁Fe_{0.1}/NF (c), Ni₁Fe_{0.3}/NF (cd) and Ni₁Fe_{0.5}/NF (e).
- **Fig. S3** SEM images of pure NF (a, b), Ni/NF (c, d), Ni₁Fe_{0.1}/NF (e, f), Ni₁Fe_{0.3}/NF (g, h) and Ni₁Fe_{0.5}/NF (g, h).

Fig. S4 SEM image (a) and XRD pattern (b) of the pretreated NF.

Fig. S5 XRD patterns of the series of electrochemical in situ grown electrodes.

- Fig. S6 (a) CV and (b) GCD curves of the series of electrodes at a scan rate of 10 mV s⁻¹ and a current density of 2 mA cm⁻², respectively. (c) CV curves of Ni₁Fe_{0.5}/NF electrode with scan rates from 2 to 50 mV s⁻¹, (d) GCD curves of Ni₁Fe_{0.5}/NF electrode with current densities from 2 to 10 mA cm⁻². (e) Specific capacity of the series of electrodes at various current densities.
- Fig. S7 Series of electrode CV and GCD curves with scan rates from 2 to 50 mV s⁻¹ and current density from 2 to 10 mA cm⁻², (a, e) Ni/NF, (b, f) Ni₁Fe_{0.1}/NF, (c, g) Ni₁Fe_{0.3}/NF and (d, h) Ni₁Fe_{0.5}/NF.
- Fig. S8 Series of electrode CV and GCD curves with scan rates from 2 to 50 mV s⁻¹ and current density from 2 to 10 mA cm⁻², (a, d) Ni₁(Fe/Co=2/1)_{0.5}/NF, (b, e) Ni₁(Fe/Co=1/2)_{0.5}/NF and (c, f) Ni₁Co_{0.5}/NF.
- Fig. S9 (a) Photographic images of the beginning and ending of the electrochemical synthesis process for unpretreated NF, (b) SEM image, (c) XRD pattern, (d) CV and (e) GCD curves.

Fig. S10 XPS of the pretreated NiFeCo/NF.

Fig. S11 Cycling performances of the series of electrodes.

- Fig. S12 SEM images of (a) before cycling test, and (b) after cycling test, with (c) TEM image and (d) SAED pattern of after cycling test.
- Fig. S13 Negative electrode (a) CV curves with the scan rates from 2 to 100 mV s⁻¹, and (b) GCD curves with the current density from 1 to 10 mA cm⁻².
- Fig. S14 CV curves of Ni₁(Fe/Co=1/1)_{0.5}/NF and AC electrodes at the scan rate of 10 mV s⁻¹.
- **Fig. S15** Photographic image of Ni₁(Fe/Co=1/1)_{0.5}/NF //AC HSC device driving LED lamp combination.
- **Video. S1** A typical video recording process of in situ electrochemical synthesis of transition metal-based hydroxides on NF (Ni₁(Fe/Co=1/1)_{0.5}/NF).
- Table. S1 Comparison of electrochemical performance of related literature ontransitionmetal-basedhydroxideselectrodes.



Fig. S1 Photographic images of the beginning and ending of the electrochemical synthesis process, and of pure NF, NF after etching treatment, and NF after electrochemical synthesis.



Fig. S2 Photographic images of pre-treatment pure NF (a), Ni/NF (b), Ni₁Fe_{0.1}/NF (c),

 $Ni_1Fe_{0.3}/NF$ (cd) and $Ni_1Fe_{0.5}/NF$ (e).



Fig. S3 SEM images of pure NF (a, b), Ni/NF (c, d), Ni₁Fe_{0.1}/NF (e, f), Ni₁Fe_{0.3}/NF (g,

h) and	Ni ₁ Fe _{0.5} /NF	(g,	h)
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Fig. S4 SEM image (a) and XRD pattern (b) of the pretreated NF.



Fig. S5 XRD patterns of the series of electrochemical in situ grown electrodes.



Fig. S6 (a) CV and (b) GCD curves of the series of electrodes at a scan rate of 10 mV s⁻¹ and a current density of 2 mA cm⁻², respectively. (c) CV curves of Ni₁Fe_{0.5}/NF electrode with scan rates from 2 to 50 mV s⁻¹, (b) GCD curves of Ni₁Fe_{0.5}/NF electrode with current densities from 2 to 10 mA cm⁻². (e) Specific capacity of the series of electrodes at various current densities.



Fig. S7 Series of electrode CV and GCD curves with scan rates from 2 to 50 mV s⁻¹ and current density from 2 to 10 mA cm⁻², (a, e) Ni/NF, (b, f) Ni₁Fe_{0.1}/NF, (c, g) Ni₁Fe_{0.3}/NF and (d, h) Ni₁Fe_{0.5}/NF.



Fig. S8 Series of electrode CV and GCD curves with scan rates from 2 to 50 mV s⁻¹ and current density from 2 to 10 mA cm⁻², (a, d) $Ni_1(Fe/Co=2/1)_{0.5}/NF$, (b, e)

 $Ni_1(Fe/Co{=}1/2)_{0.5}/NF$ and (c, f) $Ni_1Co_{0.5}/NF.$



Fig. S9 (a) Photographic images of the beginning and ending of the electrochemical synthesis process for unpretreated NF, (b) SEM image, (c) XRD pattern, (d) CV and (e) GCD curves.



Fig. S10 XPS of the pretreated NiFeCo/NF.



Fig. S11 Cycling performances of the series of electrodes.



Fig. S12 SEM images of (a) before cycling test, and (b) after cycling test, with (c) TEMimageand(d)SAEDpatternofaftercyclingtest.



Fig. S13 Negative electrode (a) CV curves with the scan rates from 2 to 100 mV s⁻¹, and (b) GCD curves with the current density from 1 to 10 mA cm⁻².



Fig. S14 CV curves of Ni_1 (Fe/Co=1/1)_{0.5}/NF and AC electrodes at the scan rate of 10

 mV

 s^{-1} .



Fig. S15 Photographic image of Ni₁(Fe/Co=1/1)_{0.5}/NF //AC HSC device driving LED lamp combination.



Video. S1 A typical video recording process of in situ electrochemical synthesis of transition

metal-based	hydroxides	on	NF	$(Ni_1(Fe/Co=1/1)_{0.5}/NF).$
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Electrode	Technical	Specific	Current	Ref.
materials	route/synthesis	canacitance	density	
matorials	time	cupucitance	density	
	time			
NC LDH NSs@Ag@CC	Two-step	1111.7 F cm ⁻²	2 mA cm^{-2}	[1]
	method/High	(502.9 mC cm ⁻²)		
	temperature for a			
	long time			
Co ₉ S ₈ /PPy/NiCo-LDH	Three-step	2.65 F cm ⁻²	1 mA cm ⁻²	[2]
NTAs	method/19 h	(1325 mC cm ⁻²)		
NiCo-LDH/Mn ₃ O ₄	Two-step	1.86 C cm ⁻²	1 mA cm ⁻²	[3]
	method/25 min			
H-NiCo LDH/ACC	Two-step	1377 mC cm ⁻²	1 mA cm ⁻²	[4]
	method/25 h			
NiO@MnO ₂	Two-step	119.4 F cm ⁻²	2 mA cm ⁻²	[5]
	method/26 h	(83.6 mC cm ⁻²)		
Co-Ni LDH–CC–CNT	Threestep	1979.2 F cm ⁻²	1 mA cm ⁻²	[6]
	method/3 h	(1187.5 mC cm ⁻²)		
Ni(OH) ₂ NTAs	Two-step	315 F cm ⁻²	18 mA	[7]
	method/5 h	(157.5 mC cm ⁻²)	cm ⁻²	
Ni ₁ (Fe/Co=1/1) _{0.5} /NF	One-step	2.32 C cm ⁻²	2 mA cm ⁻²	This
	method/400 s			work

Table. S1 Comparison of electrochemical performance of related literature on

transition metal-based hydroxides electrodes.

Note: The purpose of the technical route/synthesis time listed in Table. S1 is to reflect efficiency of the strategy proposed in this work. In particular, the technical route refers to the steps required for the synthesis of materials reported in the literature, and the synthesis time refers to the sum of time spent in the synthesis stage of materials.

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