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# Supporting Information

## A new architecture design of Ni-Co LDHs-based pseudocapacitors

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### **EXPERIMENTAL**

**Chemicals.** Nickel(II) nitrate (Ni(NO<sub>3</sub>)<sub>2</sub>·6H<sub>2</sub>O, 98%), cobalt(II) nitrate (Co(NO<sub>3</sub>)<sub>2</sub>·6H<sub>2</sub>O, 98%), and potassium hydroxide (KOH, 99%) were obtained from fisher scientific. All electrolyte solutions were prepared by Milli-Q ultrapure water (18 M $\Omega$  cm) unless stated otherwise and all the chemicals were used without any further purification.

**Procedures.** *Preparation of Cu nanowires (Cu NWs) network on copper foam.* The Cu NWs network was obtained using a standard three-electrode electrochemical cell at room temperature. Firstly, a constant anodic current density of 20 mA/cm<sup>2</sup> was applied to a cleaned copper foam  $(0.5 \times 1 \text{ cm}^2)$  for 30 minutes in 3 M freshly prepared NaOH aqueous solution at room temperature. After cleaning with deionized water, the

anodized Cu foam was reduced by applying a constant cathodic current density of 20  $mA/cm^2$  in a 0.5 M Na<sub>2</sub>SO<sub>4</sub> aqueous solution for 10 minutes.

*Fabrication of Ni-Co LDHs on Cu nanowires network.* Ni-Co LDHs were electrochemically deposited on Cu NWs in a standard three-electrode electrochemical cell at room temperature. During electrochemical deposition, the as-prepared Cu NWs network on copper foam was used as the working electrode, and the saturated calomel electrode (SCE) and a Pt foil were used as the reference electrode and counter electrode, respectively. The electrolyte solution was composed of 0.2 M Co(NO<sub>3</sub>)<sub>2</sub> and 0.1 M Ni(NO<sub>3</sub>)<sub>2</sub>. A constant cathodic current of 2.5 mA/cm<sup>2</sup> was applied to the working electrode. The deposition time was varied from 100 s to 800 s to optimize the morphology and electrochemical properties of the hybrid electrode.

The Ni-Co LDHs materials made from different ratios of Ni: Co were also synthesized by controlling the ratio of Ni: Co in solution. For clarification, we named these LDHs materials as 1-2 Ni-Co LDHs (Ni: Co = 1: 2, synthesized from 0.1 M Ni(NO<sub>3</sub>)<sub>2</sub> and 0.2 M Co(NO<sub>3</sub>)<sub>2</sub>), 1-1 Ni-Co LDHs (Ni: Co = 1: 1, synthesized from 0.15 M Ni(NO<sub>3</sub>)<sub>2</sub> and 0.15 M Co(NO<sub>3</sub>)<sub>2</sub>), 2-1 Ni-Co LDHs (Ni: Co = 2: 1, synthesized from 0.2 M Ni(NO<sub>3</sub>)<sub>2</sub> and 0.1 M Co(NO<sub>3</sub>)<sub>2</sub>), 4-1 Ni-Co LDHs (Ni: Co = 4: 1, synthesized from 0.24 M Ni(NO<sub>3</sub>)<sub>2</sub> and 0.06 M Co(NO<sub>3</sub>)<sub>2</sub>), respectively.

For comparison, nickel hydroxide or cobalt hydroxide-coated electrodes were also prepared using a similar procedure. For these electrodes,  $0.3 \text{ M Ni}(\text{NO}_3)_2$  or 0.3 M $\text{Co}(\text{NO}_3)_2$  was used as the metal ion source for the preparation of Ni hydroxide or Co hydroxide, respectively.

#### **Materials Characterization**

Scanning electron microscope (SEM) images, energy dispersive X-ray analysis (EDX) data and EDX mapping images were obtained at Hitachi S-4800 (Hitachi, Japan). Powder X-ray diffraction (XRD) was measured by Bruker Foucs D8 via ceramic monochromatized Cu Ka radiation of 1.54178 Å, operating at 40 kV and 40 mA. The scanning rate was 5° per min in  $2\theta$ . X-ray photoelectron spectroscopy (XPS) for elemental analysis was conducted on a Kratos Axis Ultra DLD X-ray Photoelectron Spectrometer. The functional groups in the prepared sample were analyzed by Fourier transform infrared (FT-IR) spectroscopy (Nicolet is10). The atomic ratio of nickel and cobalt in the Ni-Co LDHs electrode was determined by the (inductively coupled plasma optical emission spectroscopy) (ICP-OES) using the Perkin Elmer ICP-OES Optima 8300. The hybrid film was dissolved in 0.1 M HNO<sub>3</sub> with sonication and the solution was diluted to 50 mL for the ICP-OES analysis. The nickel and cobalt concentrations were obtained by using the working curve method. The N<sub>2</sub> adsorption/desorption isotherm was carried out by BET3020 (TRISTAR 3020) with the total mass of the active material (Ni-Co LDHs) and the substrate (Cu nanowires and copper foam). Electrochemical measurements were performed on a CHI 660E electrochemical workstation (Chenhua Corp., Shanghai, China). The threeelectrode system consisted of a working electrode, a platinum foil counter electrode, and a saturated calomel reference electrode (SCE, ~0.244 V vs. NHE).

Electrochemical Measurements. Electrochemical properties of the electrode material tested in a three-electrode cell system. The cyclic voltammetry (CV),

galvanostatic charge-discharge (GCD) curves and electrochemical impedance spectroscopy (EIS) of the samples were measured using a CHI 660B workstation. The EIS of the working electrode was obtained in the frequency range from 100 KHz to 0.01 Hz with an AC potential amplitude of 5 mV at zero DC voltage.

The specific capacitance (*Cs*, F/g) and the areal specific capacitance (*Ca*,  $F/cm^2$ ) of the electrode were calculated by the measured galvanostatic charge-discharge (GCD) plots according to eq S1 and S2:<sup>[1,2]</sup>

$$Cs = \frac{I \Delta t}{m \Delta V}$$
(1)  
$$Ca = \frac{I \Delta t}{s \Delta V}$$
(2)

Where I (in A) is the discharge current density,  $\Delta t$  (in s) is the discharge time, m (in g) is the total mass of the active material, s (in cm<sup>2</sup>) is the active surface area, and  $\Delta V$  (in V) is the potential window applied to the electrode.

Assembly of the hybrid supercapacitor (HSC). To determine the energy and power density of as-prepared Ni-Co LDHs materials, HSC device is prepared by using Ni-Co LDHs as a positive electrode and active carbon as a negative electrode. The electrochemical properties of the HSC were tested in 1 M KOH electrolyte. The negative electrode was fabricated by mingling active carbon, carbon black, and PVDF with a mass ratio of 8:1:1 in alcohol. This was followed by casting the mixture on a piece of Ni foam ( $1 \times 1$  cm<sup>2</sup>). After being dried in a vacuum oven at 90 °C for 6 h, the obtained material were further pressed under 10 MPa pressure before being used. To obtain a stable HSC cell operated over a wide potential window, the mass ratio of the active materials in the positive electrode and negative electrode was decided according to the charge storage balance rule  $(q^+ = q^-)$  as shows in eq S3:

$$M_{+} = M_{-} \times \frac{C_{-} \times \Delta V_{-}}{C_{+} \times \Delta V_{+}}$$
(3)

where M is mass loading of active materials, *C* is the specific capacitance obtained by testing with the three-electrodes system, and  $\Delta V$  is the voltage window of each electrode, respectively. The specific capacitance of the HSC was caculated using eq S1 on the basis of the total mass loading of positive and negative active materials. Under these conditions, the masses of Ni-Co LDHs materials and active carbon were around 0.23 mg and 18.4 mg, respectively. The energy density (*E*, Wh/Kg) and power density (*P*, W/Kg) of the HSC were caculated according to eq S4 and S5:

$$E = 0.5 C \Delta V^2 \qquad (4)$$
$$P = E/\Delta t \qquad (5)$$

where  $\Delta V$  (V) and  $\Delta t$  (s) are the working potential window and the discharge time of the HSC device.



Figure S1. The photographic images of (A) Cu foam (CF), (B) Cu(OH)<sub>2</sub> NWs/CF, (C)

Cu NWs/CF, and (D) Ni-Co LDHs/Cu NWs/CF.



**Figure S2.** N<sub>2</sub> adsorption/desorption isotherms and pore size distribution plot (insert) of the Ni-Co LDHs material.



Figure S3. SEM images of Ni-Co LDHs at different electrodeposition times: (A) 100

s, (B) 300 s, (C) 500 s and (D) 800 s.



Figure S4. CV curves (A), GCD curves (B), specific capacitance (C), and EIS (D) of

Ni-Co LDHs prepared with different electrodeposition times.



Figure S5. XRD pattern of Ni-Co LDHs scraped from the substrate.



Figure S6. High-resolution O 1s XPS of the Ni-Co LDHs/Cu NWs.



**Figure S7.** CV curves (A), GCD curves (B), calculated specific capacitance (C), and EIS (D) of Ni-Co LDHs on different substrates.



Figure S8. Rate capacitance of LDHs on Cu NWs and Cu(OH)<sub>2</sub> NWs.



Figure S9. CV curves of Cu NWs substrate and Ni-Co LDHs/Cu NWs.



**Figure S10.** (A) CV curves of the Co hydroxide electrode at various scan rates of 2 - 15 mV/s, and (B) corresponding GCD curves of the Co hydroxide electrode at different current density of 1 - 6 A/g; (C) CV curves of the Ni hydroxide electrode at various scan rates of 2 - 10 mV/s, and (D) corresponding GCD curves of the Ni hydroxide electrode at different current density of 1 - 8 A/g.



**Figure S11.** SEM images of Ni-Co LDHs prepared with different Ni: Co ratios in the precursor solution. (A) Ni: Co =1: 2; (B) Ni: Co = 1: 1; (C) Ni: Co = 2: 1, and (D) Ni: Co = 4: 1.



**Figure S12.** The electrochemical performance of the AC negative electrode in 1 M KOH electrolyte. (A) cycle voltammetry; (B) galvanostatic charge-discharge; (C) specific capacitance *vs.* current density; (D) cycling stability.



**Figure S13.** The cyclic voltammetric profiles of Ni-Co LDHs (as the positive electrode) and active carbon (as the negative electrode) at the scan rate of 5 mV/s in 1 M KOH solution.



**Figure S14.** (A) EIS of the Ni-Co LDHs//AC HSC device and (B) cycling performance of the HSC device at a current density of 2 A/g in 1 M KOH solution.

Material	Specific capacitance	Current density	Electrolyte	Ref.
Ni-Co LDHs/ Flexible carbon fiber cloth	2242.9 F/g	1 A/g	2 М КОН	[2]
Ni-Co LDHs/ textile fibers	2105 F/g	2 A/g	1 M KOH	[3]
Ni-Co-Al LDHs/ RGO	1544 F/g	1 A/g	6 M KOH	[4]
Ni-Co LDHs/ Carbon nanotube	1843 F/g	0.5 A/g	6 М КОН	[5]
Ni-Co LDHs/ Ni foam	2682 F/g	3 A/g	1 M KOH	[6]
Ni-Co LDHs/ RGO Ni foam	1054 F/g	2.5mA/cm <sup>2</sup>	6 M KOH	[7]
Ni-Co LDHs/ Ni foam	2380 F/g	1 A/g	6 M KOH	[8]
Ni-Co LDHs/ Carbon nanofiber	1949.5 F/g	1 A/g	6 М КОН	[9]
Ni-Co LDHs/ Cu NWs/CF	<b>2170</b> F/g	1 A/g	1 М КОН	Our work

**Table S1.** Comparison of specific capacitance of our Ni-Co LDHs electrode with

 other Ni-Co LDHs electrodes reported in literatures.

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