Supporting Information

# Self-powered Electrochemical Deposition of Cu@Ni(OH)<sub>2</sub> Nanobelts for High Performance Pseudocapacitors

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### **Experimental Section**

## Chemicals

CuCl<sub>2</sub>·2H<sub>2</sub>O (99.999%) and HNO<sub>3</sub> (75%) were purchased from Sigma-Aldrich. Comercial carbon conductive ink (Acheson ELECTRODAG PF-407C). Cetyltrimethylammonium chloride (CTAC) was kindly donated by Taiwan Surfactant. NiSO<sub>4</sub>·5H<sub>2</sub>O (98%) and CoSO<sub>4</sub>·7H<sub>2</sub>O (98%) were supplied by Alfa Aesar.

#### Preparation of carbon/aluminum substrate

Double-sided carbon/aluminum (C/Al) electrodes were prepared by first cleaning a piece of Al foil of 14  $\mu$ m thickness with alcohol. Comercial carbon conductive ink, which consists of finely divided carbon particles dispersed in a thermoplastic resin (Acheson ELECTRODAG PF-407C), was then coated onto the Al foil to a thickness of approximately 50  $\mu$ m. Following coating, the electrodes were dried at 90 °C for 1 h in an oven to ensure that the solvent was evaporated. The C/Al substrate was further cut into smaller circle electrodes with a diameter of 14 mm.

#### Preparation of Cu@Ni(OH)<sub>2</sub> nanobelts

As a standard procedure, 2.5 mmol of  $CuCl_2 \cdot 2H_2O$  was dissolved in 500 mL of deionized water containing 5 mM cetyltrimethylammonium chloride (CTAC) and 5 mM HNO<sub>3</sub> to obtain a limpid aqueous solution. The prepared C/Al substrate was immersed into a 10 mL of the prepared growth mediated with the carbon side facing upward in a glass vial at 290 K without stirring. Only the carbon surface gradually turned a dark-reddish color as the reaction proceeded and the Al side remained bright. After 18 h, the substrate was removed from the reaction medium, rinsed by deionized water and alcohol, and finally dried in an oven at 50°C for 30 min. The as prepared CuNBs electrode was again immersed into 5 mL of 5 mM aqueous NiSO<sub>4</sub>·5H<sub>2</sub>O without stirring. Following the growth of nickel hydroxide for 45 min, the substrates were rinsed by deionized water and alcohol, followed by drying in an oven at 50°C for 1 h. To prepare Cu@Co<sub>x</sub>Ni<sub>1</sub>.  $_{x}(OH)_{2}$  nanobelts (x= 0.2 and 0.4), 5 mL aqueous solution contained different molar ratios of CoSO<sub>4</sub>·7H<sub>2</sub>O and NiSO<sub>4</sub>·5H<sub>2</sub>O were used. For example, The synthesis of Cu@Co<sub>0.2</sub>Ni<sub>0.8</sub>(OH)<sub>2</sub> nanobelts, the growth solution comprise 1 mM CoSO<sub>4</sub>·7H<sub>2</sub>O and 4 mM NiSO<sub>4</sub>·5H<sub>2</sub>O.

#### **Material characterizations**

Scanning electron microscopic (SEM) images were obtained from a JEOL JSM-6500F at 15 kV, which was equipped with an energy dispersive spectrometer (EDS, Oxford Link Pentafet). Transmission electron microscopic (TEM) images and selected-area electron diffraction (SAED) patterns were taken from a JEOL JEM-2010F (200 kV). Powder X-ray diffraction (XRD) was performed X-ray using powder XRD (Brucker D8-advanced) using Cu- $K_{\alpha}$  radiation ( $\lambda$ =1.5405981 Å). X-ray photoelectron spectroscopy (XPS) was conducted on a PHI Quantera Spectrometer (ULVAC-PHI).

#### **Electrochemical evaluation**

Electrochemical measurements were performed by a CHI 440D instrument. The three-electrode system includes Hg/HgO as the reference electrode, platinum as the counter electrode, and Cu@Co<sub>x</sub>Ni<sub>1</sub>. <sub>x</sub>(OH)<sub>2</sub> NBs as the working electrode. 1 M KOH solution was used as the electrolyte. CV experiments were performed at a scanning rate of 5 mV s<sup>-1</sup>. GV charge/discharge curves were obtained at various current densities ranging from 10 - 100 A g<sup>-1</sup>. EIS data were recorded in a frequency range of  $0.1 - 10^5$  Hz and amplitude of 5 mV.

The specific capacitance  $(C_{sp})$  value of specimen was calculated based on the following equation:

$$C_{sp} = I \Delta t / m \Delta V$$

where *I* denotes the discharge current density (A g<sup>-1</sup>);  $\Delta V$  represents the potential window (V); and  $\Delta t$  refers to the discharge time (s).



Fig. S1 X-ray diffraction of Cu@Ni(OH)<sub>2</sub> NBs.



Fig. S2 (a) SEM images of CuNBs. (b) An energy dispersive spectrum of CuNBs.



Fig. S3 (a) Energy dispersive spectrum of Cu@Ni(OH)<sub>2</sub> NBs. (b) Plots of loaded Ni(OH)<sub>2</sub> masses (µg)

vs. time for the electrochemical deposition.



Fig. S4 TEM images, SAED patterns, and high-resolution TEM image of Cu@Ni(OH)<sub>2</sub> NBs.



Fig. S5 Comparison of the Nyquist plots made with Cu@Ni(OH)<sub>2</sub> NBs and Cu@Co<sub>x</sub>Ni<sub>1-x</sub>(OH)<sub>2</sub> NBs

(x=0.2 and 0.4) after 250 cycling.

| Electrode Material                        | Specific Capacitance <sup>a</sup>               | Rate capacity                 | Cycling fading <sup>b</sup>               | Reference |
|---|---|-------------------------------|---|-----------|
| Cu@Ni(OH) <sub>2</sub> NBs                | 2426 F g <sup>-1</sup> (10 A g <sup>-1</sup> )  | 81 % (100 A g <sup>-1</sup> ) | 72 % (30 A g <sup>-1</sup> )              | This work |
| Ni(OH) <sub>2</sub> nanospheres           | 1868 F g <sup>-1</sup> (20 A g <sup>-1</sup> )  | 71 % (70 A g <sup>-1</sup> )  | 90 % (100 mV s <sup>-1</sup> )            | 26        |
| Ni(OH) <sub>2</sub> /graphite foam        | 1650 F g <sup>-1</sup> (0.5 A g <sup>-1</sup> ) | 70 % (10 A g <sup>-1</sup> )  | 65 % (10 A g <sup>-1</sup> )              | 12        |
| Ni(OH) <sub>2</sub> /graphene<br>hydrogel | 3300 F $g^{-1}$ (0.5 A $g^{-1}$ )               | 51 % (40 A g <sup>-1</sup> )  | 95 % (100 mV s <sup>-1</sup> )            | 27        |
| Ni(OH) <sub>2</sub> /ZnO NWs              | 2226 F $g^{-1}$ (3 A $g^{-1}$ )                 | 47 % (100 A g <sup>-1</sup> ) | $36 \% (10 \text{ A g}^{-1}) ^{\text{c}}$ | 28        |
| Ni(OH) <sub>2</sub> /graphene/CNTs        | 2000 F $g^{-1}$ (0.5 A $g^{-1}$ )               | 40 % (20 A g <sup>-1</sup> )  | 50 % (10 A g <sup>-1</sup> )              | 17        |
| Ni(OH) <sub>2</sub> /Ni foam              | 3152 F g <sup>-1</sup> (4 A g <sup>-1</sup> )   | 8 % (16 A g <sup>-1</sup> )   | 48 % (4 A g <sup>-1</sup> )               | 29        |

Table S1. Comparison of the pseudocapacitor performances base on Ni(OH)<sub>2</sub>.

<sup>a</sup> measured at different current densities by GV charge/discharge test in KOH<sub>(aq)</sub>.

<sup>b</sup> capacitance fading after 1000 cycle (charging-discharging rate).

<sup>*c*</sup> estimated from the reference data.

## REFERENCE

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