Electronic Supplementary Information

High mass loading and high-density flower-like $NiCo_2O_4$ nanosheets on Ni foam for superior capacitance

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Experimental section

Chemicals： $Ni(NO_3)_2·6H_2O$ (AR), $Co(NO_3)_2·6H_2O$ (AR), Sodium acetate (AR), Polyethylene glycol 200 (GD). All chemicals were supplied by Sinopharm Chemical Reagent Co., Ltd. The active carbon (AC) was provided by Nanjing XFNANO Materials Tech Co., Ltd. All the reagents were used as received without further purification. The NF substrate was cut to 3 cm × 1 cm, immersed in 3 M HCl solution for 20 min and carefully cleaned by ultrasonication in deionized water and absolute ethanol subsequently.

Synthesis： In a typical synthesis process, 0.1455 g of $Ni(NO_3)_2·6H_2O$, 0.2910 g of $Co(NO_3)_2·6H_2O$, and 0.246 g anhydrous sodium acetate were dissolved in a mixed solution of 18 mL deionized water and 2 mL PEG200 with constant stirring at room temperature to form a homogeneous pink solution. Then, the above solution was transferred to a 50 mL Teflon-lined stainless-steel autoclave with a piece of pretreated NF immersed into the reaction solution. The autoclave was treated at 180 °C and maintained for 12 h before cooled in air. The samples were then taken out and rinsed with deionized water and ethanol several times. The as-prepared samples were calcined in air at 350 °C for 2 h with the ramp rate of 1 °C min^{-1}. The obtained materials were labeled as $NiCo_2O_4$@NF. The detailed synthesis process is depicted in Scheme 1(a). To calculate the mass loading on Ni foam accurately, the ICP atomic emission spectrometer was employed in the experiment. Through calculations, we obtained the mass loading of $NiCo_2O_4$ as 9.2 mg cm^{-2}.

Characterization： The morphologies and microstructures of the samples were investigated by SEM (Thermo VEG 3 TESCAN). HRTEM images were recorded by FEI Tecnai G2 F20 S-Twin system operated at 200 kV. X-ray diffraction (XRD, Rigaku Ultima IV) with Cu-Ka radiation at 35 kV was used to study the crystal structure of the as-synthesized products. Elemental mapping of the selected samples was conducted under SEM. The mass loading of $NiCo_2O_4$ on NF was studied by ICP (Thermo iCAP 7000).

Electrochemical performance： The electrochemical properties of samples for cyclic voltammetry (CV), galvanostatic charge-discharge (GCD), and electrochemical impedance spectroscopy (EIS) were studied in 6 M KOH solution and recorded on a CHI 660E (Chenhua) electrochemical workstation. EIS measurements were conducted in a frequency range of 100 kHz to 0.05 Hz at the open circuit potential with AC perturbation of 5 mV. In three-electrode systems,

the as prepared $NiCo_2O_4$@NF was directly used as the working electrode, with a platinum wire as the counter electrode and a SCE electrode as the reference electrode, respectively. The nominal area of the $NiCo_2O_4$@NF immersed into the electrolyte was controlled to be around 1 cm × 1 cm. The areal and specific capacitances of the electrodes were calculated from the chronopotentiometry curves and CV based on the following equations: [1-3]

$$C_a = \frac{It}{S\Delta V} \quad \text{* MERGEFORMAT (1)}$$

$$C_s = \frac{It}{m\Delta V} \quad \text{* MERGEFORMAT (2)}$$

where *Ca* (F cm^{-2}) is the areal capacitance, *Cs* (F g^{-1}) is the specific capacitance, *I* (A) is the discharge current, *t* (s) is the discharge time, ΔV (V) is the potential change during the discharge process, *S* (cm^2) is the geometrical area of the electrode and *m* (g) is the mass of the active materials on the electrode. For the two-electrode systems, the mass ratio between the positive and negative electrode is determined by balancing the charge stored in each electrode according to the following equation:

$$\frac{m^+}{m^-} = \frac{C_{s-}\Delta V_-}{C_{s+}\Delta V_+} \quad \text{* MERGEFORMAT (3)}$$

where C_{s+} and C_{s-} are the specific capacitances of $NiCo_2O_4$@NF and AC electrodes, respectively. m^+ and m^- are the active mass loadings of the positive and negative electrodes, respectively. And ΔV_+ and ΔV_- are the potential ranges of one scanning segment of $NiCo_2O_4$@NF and AC electrodes, respectively. The corresponding energy density (W h kg^{-1}) and power density (W kg^{-1}) of the ASC device can be determined from the following equations: [4]

$$E = \frac{1}{2}CV^2 \quad \text{* MERGEFORMAT (4)}$$

$$P = \frac{E}{\Delta t} \quad \text{* MERGEFORMAT (5)}$$

where *C* (F g^{-1}) is the specific capacitance of the ASC device tested from GCD curves, and *V* (V) is the potential range of the assembled device, Δt (s) is the discharge time obtained from the charge–discharge test.

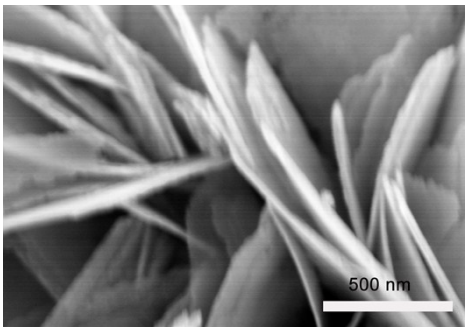

Fig S1 SEM image of flower-like $NiCo_2O_4$ nanosheets on NF at higher magnification.

Supplementary References

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3. Y. Deng, Y. Ji, H. Wu and F. Chen, *Chem. Commun.*, 2019, **55**, 1486-1489.

4. Y. Ji, Y. Deng, H. Wu and Z. Tong, *J. Appl. Electrochem.*, 2019, **49**, 599-607.