**Electronic Supplementary Inforamtion for** 

## Novel topotactically transformed carbon-CoO-NiO-NiCo<sub>2</sub>O<sub>4</sub> nanosheets hybrid hetero-structured arrays as ultrahigh performance supercapacitors

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**Preparation of NiCo<sub>x</sub>(OH)**<sub>y</sub> **NSHAs on nickel foam**. The precursor nanosheets arrays were synthesized on nickel foam through a facile hydrothermal method according to Tu's previous work.<sup>1</sup> 1 mmol of Ni(NO<sub>3</sub>)<sub>2</sub>, 2 mmol of Co(NO<sub>3</sub>)<sub>2</sub>, 6 mmol of NH<sub>4</sub>F, and 15 mmol of urea were dissolved in 70 mL of deionized water under magnetical stirring for 30 min in air. Then the obtained solution was transferred into a 90 mL Teflon-lined stainless steel autoclave and the clean substrate of nickel foam with  $2.0 \times 5.0$  cm in size was immersed into it. Then the autoclave was heated to  $120^{\circ}$ C inside an electric oven for 5 h. As soon as the reaction finished, the nickel foam with precursor was taken out and rinsed by deionized water.

**Preparation of NiCo<sub>2</sub>O<sub>4</sub> NSHAs on nickel foam**. To obtain pure NiCo<sub>2</sub>O<sub>4</sub> nanosheets arrays, the NiCo<sub>x</sub>(OH)<sub>y</sub> precursor on the nickel foam was transferred into a quartz tube furnace. In an atmosphere of N<sub>2</sub> at the flow rate of 100 sccm, the sample was heated to 350°C and held for 3 h for completely converting into NiCo<sub>2</sub>O<sub>4</sub> nanosheets arrays. Finally, the sample was cooled down to room temperature naturally with atmosphere of N<sub>2</sub> and taken out and NiCo<sub>2</sub>O<sub>4</sub> NSHAs were obtained.

**Preparation of NiCo<sub>2</sub>O<sub>4</sub>/C NSHAs on nickel foam.** In order to get heterostructured NiCo<sub>2</sub>O<sub>4</sub>/C nanosheets arrays, the NiCo<sub>x</sub>(OH)<sub>y</sub> precursor on the nickel foam was transferred into a quartz tube furnace. In an atmosphere of N<sub>2</sub> at the flow rate of 100 sccm, the sample was heated to 350°C and held for 3 h, subsequently, C<sub>2</sub>H<sub>2</sub> was introduced immediately at the flow rate of 10 sccm for 150 s with the N<sub>2</sub> unchanged all the time. Finally, the sample was cooled down to room temperature naturally with atmosphere of N<sub>2</sub> and taken out and heterostructured NiCo<sub>2</sub>O<sub>4</sub>/C NSHAs were obtained.

## **XRD** spectrum



Fig. S1 XRD patterns of NiCo<sub>2</sub>O<sub>4</sub> and NiCo<sub>2</sub>O<sub>4</sub>/C NSHAs.

## **XPS spectrum and analysis**

Fig. S2 displays the high-resolution XPS spectrum of Co 2p, Ni 2p and O 1s for NiCo<sub>2</sub>O<sub>4</sub> NSHAs in Fig. S2 (a, b, c) and Co 2p, Ni 2p and O 1s for NiCo<sub>2</sub>O<sub>4</sub>/C NSHAs in Fig. S2 (d, e, f), respectively. By using a Gaussian fitting method, the Co 2p emission spectrum (Fig. S 2b) was fitted with two spin-orbit doublets of  $Co^{2+}$  and  $Co^{3+}$  and negligible satellites (identified as Sat.). Such result is in agreement with the literature for NiCo<sub>2</sub>O<sub>4</sub>.<sup>2-3</sup> Then, after the carbon was integrated, two obvious Sat., which were only detected in CoO as shown in Fig. S2d, which convinced that CoO generated during the CVD stage.<sup>3</sup> As for Ni 2p, the results of both products are the same except for a distinguished peak at 852.8 eV, which come from Ni<sup>2+</sup>, probably NiO.<sup>4</sup> The spectrum for the O 1s region of NiCo<sub>2</sub>O<sub>4</sub> and NiCo<sub>2</sub>O<sub>4</sub> /C shows four same oxygen contributions, which have been denoted as O1, O2, O3, O4, O5. Peaks O1 and O2, O5 at around 529.5 eV are typical of the metal-oxygen bond.<sup>2,5</sup> With respect to the component O3, the fitting peak at 531.1 eV is commonly associated with defects, chemisorbed oxygen or under-coordinated lattice oxygen.<sup>5-6</sup> The component O4 can be attributed to multiplicity of physic- and chemi-sorbed water at or near the surface.<sup>2,5</sup> Such results further indicate that the partial NiCo<sub>2</sub>O<sub>4</sub> was reduced to CoO and NiO.



Fig. S2. High-resolution XPS spectrum of  $NiCo_2O_4$  NSHAs: ((a) Co 2p, (b) Ni 2p (c) O 1s) and  $NiCo_2O_4/C$  NSHAs ((d) Co 2p, (e) Ni 2p (f) O 1s).



Fig. S3 discharge curves versus various current density: (a)  $NiCo_2O_4$  NSHAs; (b)  $NiCo_2O_4/C$  NSHAs.



Fig. S4. EIS spectrum and corresponding curve fitting for NiCo<sub>2</sub>O<sub>4</sub> and NiCo<sub>2</sub>O<sub>4</sub>/C NSHAs.

**Materials characterizations.** The products were characterized using X-ray diffraction (XRD, PANalytical Empyrean, Cu K $\alpha$  radiation;  $\lambda$ =1.5418 Å), field-emission scanning electron microscopy (FESEM, JEOL JSM-6700F, 10 kV), transmission electron microscopy and elemental (TEM, Titan G2 60-300), Raman spectroscopy (LabRAM HR evolution with laser excitation at 532 nm). The mass of electrode materials was measured on an AX/MX/UMX Balance (METTLER TOLEDO, maximum =5.1 g; d =0.001 mg). To characterize the electrochemical behaviors, CHI660C (CH Instruments Inc. Shanghai) electrochemical workstation was used in a three-electrode electrochemical cell using a 6 M KOH aqueous solution as electrolyte. Electrochemical Impedance Spectroscopy was tested on CS310 by applying an AC voltage with 5 mV amplitude in a frequency range from 0.01 Hz to 100 kHz at open circuit potential.

The area specific capacitance of the electrodes was calculated from the CP curves based on following equation:

$$C = \frac{It}{\Delta V \mathbf{g} M} \square$$

C is the specific capacitance electrochemical materials (F g<sup>-1</sup>). I is the discharging current density (A cm<sup>-2</sup>), t is the discharging time (s),  $\Delta V$  is the discharging potential range (V), M is the mass including the active material except the nickel foam, respectively.

## References

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