Electronic supplementary Information for

Nano-aggregates of cobalt nickel oxysulfide as a high-performance

electrode material for supercapacitors

Lifeng Liu*

International Iberian Nanotechnology Laboratory (INL), 4715-330 Braga, Portugal

* Corresponding author. E-mail: lifeng.liu@inl.int

Experimental details

The (CoNi)O_xS_y nano-aggregates were prepared by a facile hydrothermal method. First of all, Co(NO₃)₂·6H₂O and NiSO₄·6H₂O (Sigma-Aldrich, analytical grade) were dissolved into de-ionized water (\geq 18 MΩ·cm) to form 0.01 M aqueous solution. Subsequently, 25ml mixed solution was blended with 25 ml 0.01 M Na₂S·9H₂O (Sigma-Adrich, analytical grade) drop by drop. Black precipitates immediately appeared upon blending. The as-obtained mixture was then transferred into a Teflon-lined stainless steel autoclave reactor. After that, high-purity nitrogen (99.999 %) was flowed into the reactor, resulting in an internal pressure of 6 bars. The reactor was subsequently heated to 180 °C and maintained at that temperature for 18 h, and then cooled down to room temperature. The internal pressure in the reactor maintained at around 15 bars during the whole reaction process. The resulting black powders were collected by centrifugation, washed in de-ionized water and subjected to centrifugation once again. Afterwards, the powders were dispersed into 0.5 ml de-ionized water to form a slurry. The morphology of the as-prepared nanostructures was characterized by scanning electron microscopy (SEM, FEI Quanta 650), and their microstructure was examined by transmission electron microscopy (TEM, FEI Titan ChemSTEM 80-200, probe corrected). X-ray photoelectron spectroscopy (XPS) measurement was performed using a Thermo Scientific EscaLab 250Xi system equipped with a monochoromated AI K α X-ray radiation source. All spectra were calibrated with respect to the three standard peaks, namely, Au 4f_{7/2}, Ag 3d_{5/2} and Cu2p_{3/2}. The C 1s peak was observed at 284.6 eV. The electrochemical tests were carried out in a three-electrode cell using a Pt wire as counter electrode and a saturated calomel electrode (SCE) as reference, respectively. The working electrode was prepared by spreading the (CoNi)O_xS_y slurry over a nickel foam current collector, followed by a thermal treatment in high-purity argon (99.9999%) at 250 °C for 1 h. It was found that the black powders can strongly adhere to the surface of nickel foam after annealing. The typical loading was 2.5 – 3.5 mg/cm². The cyclic voltammetry (CV) and galvanostatic charge/discharge tests were performed in a 2 M NaOH aqueous solution using a multi-channel battery tester (Biologic VMP3).

The specific and areal capacitance are calculated according to the following equations:

$$C_s = \frac{I\,\Delta t}{m\,\Delta U}\tag{1}$$

$$C_a = \frac{I \,\Delta t}{A \,\Delta U} \tag{2}$$

where C_s and C_a represent specific and areal capacitance, respectively, *I* is the charge/discharge current, Δt the discharge time, ΔU potential window, *m* the mass loading of active materials, *A* the area of the capacitor.

Supplementary tables

Table S1. The variation of specific capacitance, areal capacitance, specific energy density and specific power density as a function of charge/discharge current densities.

Current density (A g ⁻¹)	0.5	1	2	5	10	20	30	50
Current density (mA cm ⁻²)	1.375	2.75	5.5	13.75	27.5	55	82.5	137.5
Specific capacitance	592	485	381	276	215	160	127	85
(<i>C</i> _s , F g ⁻¹)								
Areal capacitance	1628	1333	1048	760	590	440	350	233
(<i>C</i> _a , mF cm ⁻²)								
Specific energy density	27.0	22.0	17.8	13.3	10.0	6.7	4.7	2.5
(Wh kg ⁻¹)								
Specific power density	136.9	271.6	562.6	1464.2	2880.0	5481.8	7356.5	10000.0
(W kg ⁻¹)								