Electronic Supporting Information

Globular Reduced Graphene Oxide /Metal Oxide Structures for Energy Storage Applications

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Preparation of rGO/ MO composites

In a typical synthesis, 9.784 g cobalt hydroxide ($Co(OH)_2$, 95%, Sigma) powder was solved in 100 ml of 1M nitric acid solution. To obtain the composites, cobalt hydroxide ($Co(OH)_2$, 95%, Aldrich) powder dissolved in 1M nitric acid solution was added into a diluted GO dispersion in water. Then the solution was stirred for 30 min using a conventional magnetic stirrer. The hybrid material was then obtained *in situ* by spraypyrolyzing the suspensions into a vertical type spray pyrolysis reactor to obtain 20% rGO/Co_3O_4 composites. A typical range of reaction temperatures is about 500 °C to about 1000 °C, with about 600 °C being preferable for a fluid flow rate of between about 4 ml min^{-1} and about 20 ml min^{-1}. Additionally, when a noble carrier gas is used, a positive partial pressure for the noble carrier gas can be introduced within the reaction chamber which influences the produced graphene oxide based composite. An example positive partial pressure that can be used is between about 3 kPa and about 10 kPa.

The same procedure was performed for the preparation of the rGO-NiO composite. This time 29.079 g crystalline Nickel (II) nitrate hexahydrate ($Ni(NO_3)_2 \cdot 6H_2O$, Sigma) powder was

dissolved in water and then added into a diluted GO dispersion in water with different mass ratios to obtain 20 % rGO/NiO .

Equipment

The working electrode was prepared by coating the active material on to stainless steel sheets (1cm × 1cm) previously polished with sand paper and ultrasonicated in ethanol for an hour. 7 mg of the electroactive material (rGOMO) were mixed with 2 mg of carbon black and 1 mg of PVDF binder in an Agate mortar in (N-methyl pyrrolidinone) NMP solvent and ground using a pestle. The resulting slurry was then spread on to the polished stainless steel surface to achieve mass loadings between 0.85 and 1 mg and allowed to dry in a vacuum oven for 24 hours. Specific capacitance calculations were performed using 70% of the total electrode mass loading since the remaining 30% of the mass is contributed by carbon black and PVDF binder that do not significantly contribute to the overall capacitance.

X-ray diffraction experiments were performed at room temperature using high resolution X-ray diffraction system (GBC MMA X-Ray diffraction (XRD) with Cu-Kα radiation). The elemental composition was characterized using the X-ray photoelectron spectroscopy (XPS, PHOIBOS 100 hemispherical analyzer produced by SPECS GmbH) with pass energy of 26.00 eV, 45° takeoff angle and a beam size of 100 µm. The morphologies of the composites were examined by scanning electron microscope (JSM 7500F, JEOL). BET measurements were performed on the Nova 1000 instrument. HRTEM was performed on the JEOL F3000) machine.

Electrochemical Characterization

Electrochemical experiments were performed at room temperature on the CHI660C (CH Instruments, Inc) electrochemical workstation using the three electrode system in a beaker type cell. An electrolyte solution of 1M NaOH, a Ag|AgCl reference electrode and a platinum foil as a

counter electrode were used. Cyclic voltammetry and was performed over a voltage range of -0.2 to 0.5 V at various scan rates (5 to 100 mVs-1). Specific capacitances were calculated at 5 mVs^{-1}. EIS measurements were carried out between 10 kHz and 0.01 Hz using a 5 mV rms sinusoidal modulation.

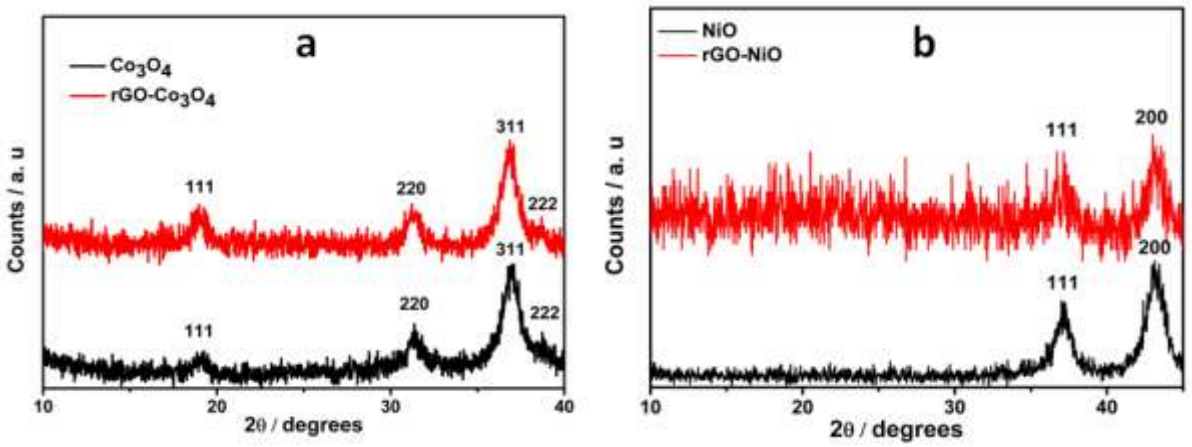

Figure S1. XRD profiles for (a) Co_3O_4 and rGO-Co_3O_4 and (b) NiO and rGO-NiO composites after spray pyrolysis.

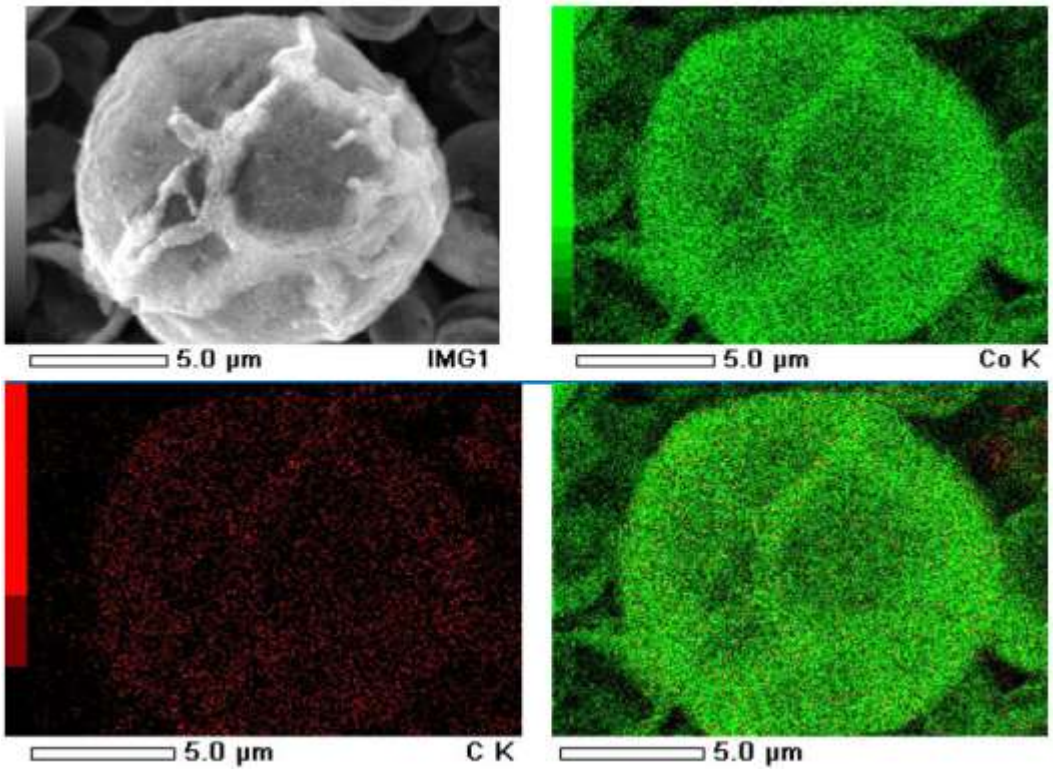

Figure S2. The EDS mapping showing good homogeneity of the obtained nanocomposite particles, demonstrated by the substantially even distribution of Co and carbon across the sample

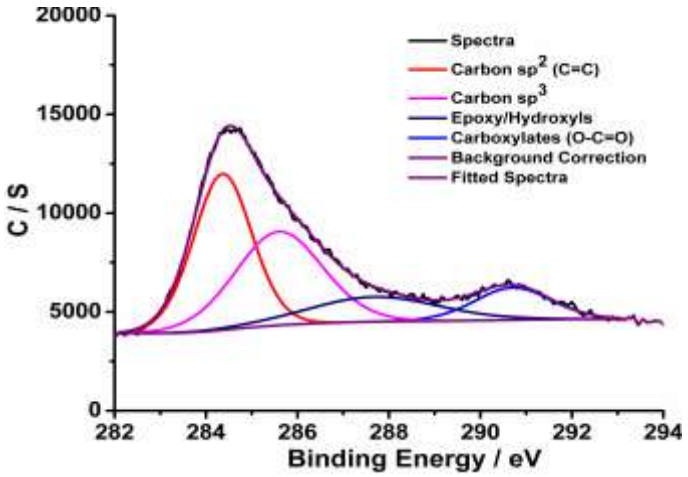

Figure S3. XPS spectra of rGO-Co_3O_4 composite after spray pyrolysis.

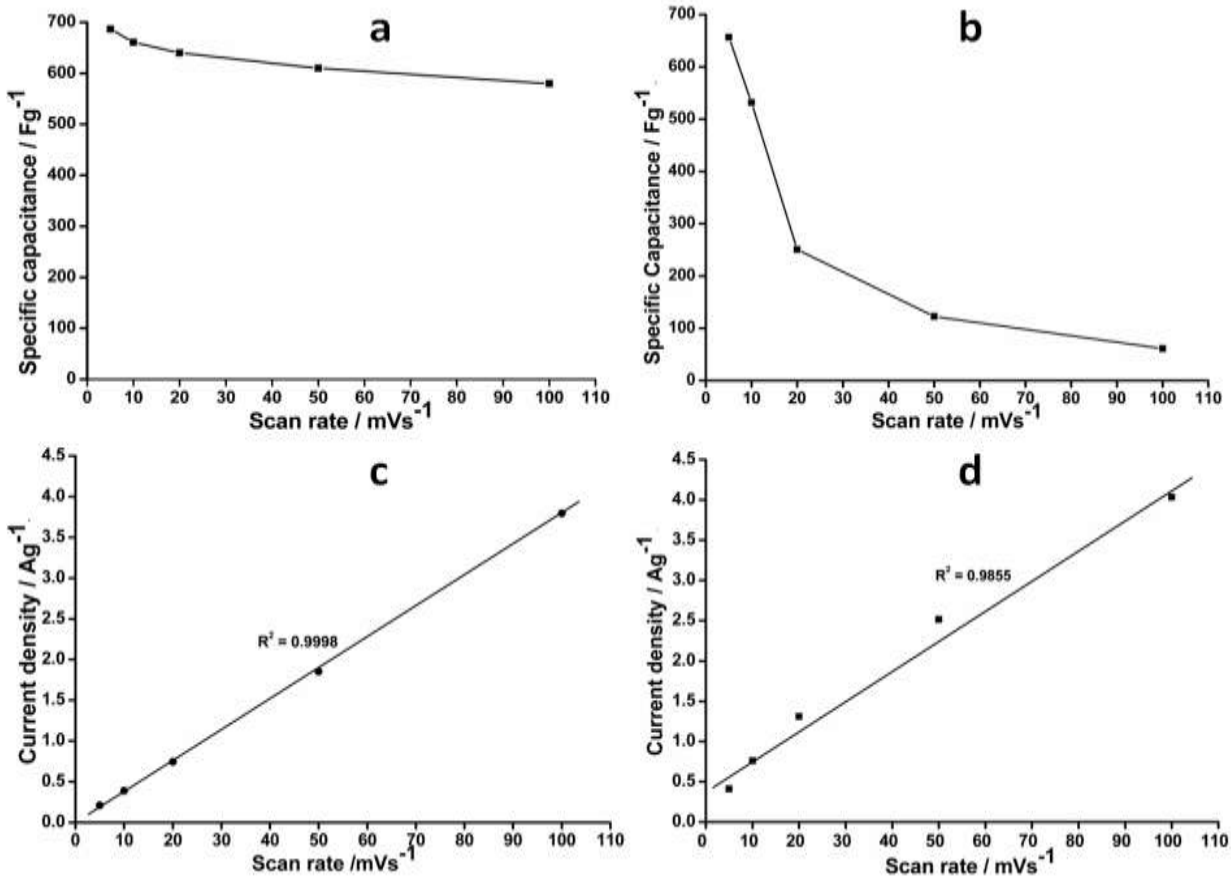

Figure S4. Variation of specific capacitance with scan rate for (a) rGO-Co_3O_4 and (b) rGO-NiO and current density with scan rate for (c) rGO-Co_3O_4 and (d) rGO-NiO.