Supporting Information

**Oxygen Defect-mediated NiCo$_2$O$_4$ Nanosheets as Electrode for Pseudocapacitor with Improved Rate Capability**

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Experimental Section

Synthesis of r-NiCo$_2$O$_4$ nanosheets: In a typical synthesis, 1.1 mmol Ni(NO$_3$)$_2$•6H$_2$O, 2.2 mmol Co(NO$_3$)$_2$•6H$_2$O and 13.2 mmol hexamethylenetetramine (HMT) are dissolved into 80 mL mixed solution of ethylene glycol/water (v/v=7/1) to form the clear pink solution after violent stirring for 30 min. Then the clear pink solution is transferred into a 100 mL Teflon lined stainless autoclave. The autoclave is sealed and maintained at 120°C for 4 hours and then cooled down to room temperature. The precipitates are collected by centrifugation and rinsed for three times with abundant amount of deionized water and ethanol, respectively. Then, the green solid product is dried at −40°C in freezing dryer for 36 h. The dried samples are further calcined at 300°C for 2 hours in the air to convert the hydroxide into the NiCo$_2$O$_4$. For oxygen defect-mediated NiCo$_2$O$_4$, 0.1 g original NiCo$_2$O$_4$ NSs are immersed in 100 mL 1.0 M NaBH$_4$ solution for 1 h at room temperature. After reduction, the samples are filtered, thoroughly washed with deionized water and ethanol for several times and dried in a vacuum oven at 60°C for 12 h. The synthesis process of the irregular NiCo$_2$O$_4$ NPs is similar to the aforementioned method, while the solvent is all deionized water and the reaction time is 12 hours.

Characterization: The samples are characterized by field emission scanning electron microscopy (FESEM, Hitachi, S-4800), transmission electron microscopy (TEM, JEOL, JEM-1101), high-resolution transmission electron microscopy (HRTEM, JEOL, JEM-2100) and X-ray powder diffractometry.
(XRD, Bruker D8 Advance X-ray diffractometer system with Co Kα radiation of 1.7902 Å.). The specific surface areas of the products are evaluated by the ASAP 2020 instrument. An X-ray photoelectron spectroscopy (XPS) survey is conducted using a PHI 5000 VersaProbe with an Al Kα excitation source. The Raman spectra are obtained using a Renishaw Via Reflex Raman system with the excitation laser of 532 nm. The electrical conductivity is measured by a four-wire method using a source measure unit (SMU, Keithley 6430). Thermal gravimetry analysis (TGA) is recorded on Netzsch STA 449C under air and nitrogen atmosphere with the heating rate of 5 °C/min.

_Electrochemical Measurements:_ The working electrode is prepared by mixing 80 wt.% as-made products, 10 wt.% acetylene black, and 10 wt.% polytetrafluoroethylene (PTFE) in small amount of ethanol to obtain the slurry mixture. The obtained slurry is coated on the single surface of nickel foams with a size of 1 cm × 1 cm and dried in vacuum at 80°C for 12 h. Then, the nickel foams with the electroactive materials are pressed under 10 MPa with the thickness of 0.11 mm. The mass loading of active material is about 1.8 mg cm⁻². The electrochemical tests of all samples are conducted in a three-electrode system with a Pt counter-electrode and an Hg/HgO reference electrode in 6.0 M KOH solution. The electrochemical AC impedance measurements are carried out to assess the electrical properties at an open circuit voltage with frequency from 0.01 Hz to 100 kHz and an amplitude of 5 mV. All the electrochemical tests are carried out on the electrochemical working station (CHI660E, Shanghai, China).
The specific capacitance of the supercapacitor can be calculated by galvanostatic discharge-charge (GCD) test as following equation:

\[ C_m = \frac{I}{m} \times \frac{\Delta t}{\Delta V} \]

Where \( C_m \) is the specific capacitance of the capacitor (F g\(^{-1}\)); \( I \) is the current of the charge/discharge process; \( \Delta t \) is the discharging time period in seconds for the potential change \( \Delta V \), in volts; \( m \) is the mass loading of the active material. All the electrochemical measurements are carried out at room temperature.
**Figure S1.** SEM images of nickel-cobalt hydroxides nanosheets obtained at various solvothermal reaction times: (a) and (a’) for 1 h; (b) and (b’) for 2 h; (c) and (c’) for 4 h; (d) and (d’) for 6 h.
Figure S2. TEM images of optimized nickel-cobalt hydroxide nanosheets at 120 °C for 4 h.
Figure S3. TGA curve of optimized nickel-cobalt hydroxide nanosheets at 120 °C for 4 h.
Figure S4. TGA analysis of (a) NiCo$_2$O$_4$ NPs, (b) original NiCo$_2$O$_4$ NSs and (c) r-NiCo$_2$O$_4$ NSs in air and argon atmosphere. (Flow of 20 mL min$^{-1}$ and a ramping rate of 5°C/min). The concentrations of oxygen defects are calculated from the difference in weight decrease between the two TGA curves.
Figure S5. CV curves of NiCo$_2$O$_4$ NPs, original NiCo$_2$O$_4$ and r-NiCo$_2$O$_4$ NSs at various scan rates.

Figure S6. (a) The polarization currents at 0.5 V and the oxidation currents of NiCo$_2$O$_4$ NPs, NiCo$_2$O$_4$ NSs, and r-NiCo$_2$O$_4$ NSs electrodes. (b) Coulombic efficiencies of NiCo$_2$O$_4$ NPs, NiCo$_2$O$_4$ NSs, and r-NiCo$_2$O$_4$ NSs at the current density of 20 A g$^{-1}$. 
Figure S7. (a-c) Galvanostatic charge and discharge voltage profiles of NiCo$_2$O$_4$ NPs, original NiCo$_2$O$_4$ and r-NiCo$_2$O$_4$ NSs at various current densities.
**Figure S8.** Plots of specific capacitance as a function of scan rate of NiCo$_2$O$_4$ NPs, original NiCo$_2$O$_4$ NSs, and r-NiCo$_2$O$_4$ NSs.
Figure S9. Diffusion and capacitive-controlled current contributions of NiCo$_2$O$_4$ NPs, original NiCo$_2$O$_4$ NSs, and r-NiCo$_2$O$_4$ NSs calculated from CV curves at the scan rate of 5 mV s$^{-1}$. 
Table S1. Comparison of the electrochemical performances of r-NiCo$_2$O$_4$ electrode in this work with NiCo$_2$O$_4$ and carbon-based electrodes in the literature

<table>
<thead>
<tr>
<th>NiCo$_2$O$_4$-based electrode</th>
<th>Specific capacitance</th>
<th>Rate capability</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$C_{\text{min}}$</td>
<td>$C_{\text{max}}$</td>
<td></td>
</tr>
<tr>
<td>Mesoporous NiCo$_2$O$_4$</td>
<td>956 F g$^{-1}$ @ 1 A g$^{-1}$</td>
<td>828 F g$^{-1}$ @ 10 A g$^{-1}$</td>
<td>86.6% (1-10 A g$^{-1}$)</td>
</tr>
<tr>
<td>rGO wrapped porous NiCo$_2$O$_4$</td>
<td>1176 F g$^{-1}$ @ 2 A g$^{-1}$</td>
<td>1020 F g$^{-1}$ @ 8 A g$^{-1}$</td>
<td>86.7% (2-8 A g$^{-1}$)</td>
</tr>
<tr>
<td>NiCo$_2$O$_4$-decorated porous carbon</td>
<td>596.8 F g$^{-1}$ @ 2 A g$^{-1}$</td>
<td>185 F g$^{-1}$ @ 20 A g$^{-1}$</td>
<td>30.9% (2-20 A g$^{-1}$)</td>
</tr>
<tr>
<td>NiCo$_2$O$_4$ tetragonal microtubes</td>
<td>1388 F g$^{-1}$ @ 2 A g$^{-1}$</td>
<td>863 F g$^{-1}$ @ 30 A g$^{-1}$</td>
<td>62.2% (2-30 A g$^{-1}$)</td>
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<tr>
<td>nickel cobalt oxide/graphene oxide</td>
<td>1211 F g$^{-1}$ @ 1 A g$^{-1}$</td>
<td>687 F g$^{-1}$ @ 10 A g$^{-1}$</td>
<td>56.7% (1-10 A g$^{-1}$)</td>
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<tr>
<td>NiCo$_2$O$_4$ hollow spheres</td>
<td>1204 F g$^{-1}$ @ 2 A g$^{-1}$</td>
<td>822 F g$^{-1}$ @ 20 A g$^{-1}$</td>
<td>68.3% (1-20 A g$^{-1}$)</td>
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<td>NiCo$_2$O$_4$ microspheres</td>
<td>1822.3 F g$^{-1}$ @ 2 A g$^{-1}$</td>
<td>1250.9 F g$^{-1}$ @ 20 A g$^{-1}$</td>
<td>68.6% (1-20 A g$^{-1}$)</td>
</tr>
<tr>
<td>Ni/Co porous oxide nanosheets</td>
<td>830 F g$^{-1}$ @ 1 A g$^{-1}$</td>
<td>710 F g$^{-1}$ @ 20 A g$^{-1}$</td>
<td>85.5% (1-20 A g$^{-1}$)</td>
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<td>Mesoporous NiCo$_2$O$_4$</td>
<td>655 F g$^{-1}$ @ 1 A g$^{-1}$</td>
<td>430 F g$^{-1}$ @ 10 A g$^{-1}$</td>
<td>65.6% (1-10 A g$^{-1}$)</td>
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<tr>
<td>hollow NiCo$_2$O$_4$ nanospheres</td>
<td>1229 F g$^{-1}$ @ 1 A g$^{-1}$</td>
<td>1027 F g$^{-1}$ @ 25 A g$^{-1}$</td>
<td>83.6% (1-25 A g$^{-1}$)</td>
</tr>
<tr>
<td>Mesoporous NiCo$_2$O$_4$/NF</td>
<td>708 F g$^{-1}$ @ 1 A g$^{-1}$</td>
<td>628 F g$^{-1}$ @ 10 A g$^{-1}$</td>
<td>88.7% (1-10 A g$^{-1}$)</td>
</tr>
<tr>
<td>N-doped graphene framework (NCO/NGF)</td>
<td>1198 F g$^{-1}$ @ 1 A g$^{-1}$</td>
<td>634 F g$^{-1}$ @ 20 A g$^{-1}$</td>
<td>52.9% (1-20 A g$^{-1}$)</td>
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<tr>
<td>rGO/NiCo$_2$O$_4$</td>
<td>1003 F g$^{-1}$ @ 1 A g$^{-1}$</td>
<td>893 F g$^{-1}$ @ 10 A g$^{-1}$</td>
<td>89.0% (1-10 A g$^{-1}$)</td>
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<tr>
<td>NiCo$_2$O$_4$/CuO-x</td>
<td>1670 F g$^{-1}$ @ 1 A g$^{-1}$</td>
<td>692 F g$^{-1}$ @ 10 A g$^{-1}$</td>
<td>41.4% (1-10 A g$^{-1}$)</td>
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<tr>
<td>rHGO/ NiCo$_2$O$_4$@CF</td>
<td>1178 F g$^{-1}$ @ 1 A g$^{-1}$</td>
<td>1100.4 F g$^{-1}$ @ 10 A g$^{-1}$</td>
<td>93.4% (1-10 A g$^{-1}$)</td>
</tr>
<tr>
<td>Material/Modification</td>
<td>Specific Capacity @ 1 A g(^{-1}) (F g(^{-1}))</td>
<td>Specific Capacity @ 10 A g(^{-1}) (F g(^{-1}))</td>
<td>Yield (1-10 A g(^{-1}))</td>
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</tr>
<tr>
<td>Co–Ni–O/3DG</td>
<td>1586 F g(^{-1})</td>
<td>~1100 F g(^{-1})</td>
<td>69.4% (1-10 A g(^{-1}))</td>
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<tr>
<td>NiCo(_2)O(_4@)GQDs</td>
<td>1382 F g(^{-1})</td>
<td>1278 F g(^{-1})</td>
<td>92.5% (1-20 A g(^{-1}))</td>
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<td>hexagonal NiCo(_2)O(_4)</td>
<td>1525 F g(^{-1})</td>
<td>610 F g(^{-1})</td>
<td>40.0% (1-10 A g(^{-1}))</td>
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<tr>
<td>NiCo(_2)O(_4) Nanoparticle</td>
<td>1084 F g(^{-1})</td>
<td>300 F g(^{-1})</td>
<td>27.7% (2-10 A g(^{-1}))</td>
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<tr>
<td>P- NiCo(_2)O(_4)</td>
<td>1642 F g(^{-1})</td>
<td>1335 F g(^{-1})</td>
<td>81.3% (2-20 A g(^{-1}))</td>
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<tr>
<td>NiCo(_2)O(_4@)AMCRs</td>
<td>1691 F g(^{-1})</td>
<td>1275 F g(^{-1})</td>
<td>75.4% (3-20 A g(^{-1}))</td>
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<tr>
<td>NiCo(_2)O(_4/PANI/MF)</td>
<td>1540.1 F g(^{-1})</td>
<td>1220 F g(^{-1})</td>
<td>79.2% (1-20 A g(^{-1}))</td>
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<tr>
<td>Mesoporous NiCo(_2)O(_4) flower</td>
<td>122.5 C g(^{-1})</td>
<td>82.5 C g(^{-1})</td>
<td>67.3% (1-10 A g(^{-1}))</td>
</tr>
<tr>
<td>NiCo(_2)O(_4/)carbon</td>
<td>1480.9 F g(^{-1})</td>
<td>995.2 F g(^{-1})</td>
<td>67.2% (1-20 A g(^{-1}))</td>
</tr>
<tr>
<td>NiCo(_2)O(_4/)Carbonized melamine foam</td>
<td>1541 F g(^{-1})</td>
<td>1120 F g(^{-1})</td>
<td>72.7% (1-20 A g(^{-1}))</td>
</tr>
<tr>
<td>Hierarchically porous carbon</td>
<td>218.6 F g(^{-1})</td>
<td>164.5 F g(^{-1})</td>
<td>75.3% (1-20 A g(^{-1}))</td>
</tr>
<tr>
<td>O-N-S co-doped hierarchical porous carbon</td>
<td>576 F g(^{-1})</td>
<td>267 F g(^{-1})</td>
<td>43.4% (1-20 A g(^{-1}))</td>
</tr>
<tr>
<td>Porous carbon sheets</td>
<td>407 F g(^{-1})</td>
<td>224 F g(^{-1})</td>
<td>55.0% (1-20 A g(^{-1}))</td>
</tr>
<tr>
<td>Carbon spheres/porous carbon</td>
<td>337 F g(^{-1})</td>
<td>280 F g(^{-1})</td>
<td>83.1% (1-20 A g(^{-1}))</td>
</tr>
<tr>
<td>N-doped porous graphene</td>
<td>390 F g(^{-1})</td>
<td>238 F g(^{-1})</td>
<td>61.0% (1-20 A g(^{-1}))</td>
</tr>
<tr>
<td>Ultrathin NiCo(_2)O(_4) Nanosheets/NF</td>
<td>2010 F g(^{-1})</td>
<td>1450 F g(^{-1})</td>
<td>72.1% (2-20 A g(^{-1}))</td>
</tr>
<tr>
<td>Ultrathin NiCo(_2)O(_4) Nanosheets/CF</td>
<td>1002 F g(^{-1})</td>
<td>520 F g(^{-1})</td>
<td>48.8% (1-20 A g(^{-1}))</td>
</tr>
<tr>
<td>Porous NiCo(_2)O(_4) Nanosheet/Carbon Fabric</td>
<td>2658 F g(^{-1})</td>
<td>1866 F g(^{-1})</td>
<td>70.2% (2-20 A g(^{-1}))</td>
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<tr>
<td>Pd-NiCo(_2)O(_4)/NF</td>
<td>2484.4 F g(^{-1})</td>
<td>2011 F g(^{-1})</td>
<td>80.9% (2-20 A g(^{-1}))</td>
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<tr>
<td>Mesoporous NiCo(_2)O(_4) Nanosheets</td>
<td>560 F g(^{-1})</td>
<td>400 F g(^{-1})</td>
<td>71.4% (2-20 A g(^{-1}))</td>
</tr>
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<td>r-NiCo(_2)O(_4) nanosheets</td>
<td>1980 F g(^{-1})</td>
<td>1897 F g(^{-1})</td>
<td>95.8% (1-10 A g(^{-1}))</td>
</tr>
</tbody>
</table>
Reference


[21] W. Jiang, F. Hu, Q. Yan, X. Wu, Investigation on electrochemical behaviors of NiCo$_2$O$_4$ battery-type supercapacitor electrodes: the role of


