Supporting Information

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A Universal Strategy to Prepare Functional Porous Graphene Hybrid Architectures

Zhiqiang Niu, Lili Liu, Li Zhang, Qi Shao, Weiya Zhou, Xiaodong Chen*, and Sishen Xie
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By Zhiqiang Niu, Lili Liu, Li Zhang, Qi Shao, Weiya Zhou, Xiaodong Chen* and Sishen Xie

[*] Prof. X. D. Chen, Dr. Z. Q. Niu, Dr. L. L. Liu, Dr. L. Zhang and Q. Shao
School of Materials Science and Engineering, Nanyang Technological University, 50 Nanyang Avenue, Singapore 639798
E-mail: chenxd@ntu.edu.sg
Website: http://www.ntu.edu.sg/home/chenxd/

Prof. W. Y. Zhou, Prof. S. S. Xie
Beijing National Laboratory for Condensed Matter Physics, Institute of Physics, Chinese Academy of Sciences, Beijing 100190, P. R. China

[+ ] Z. Niu and L. Liu contributed equally to this work

Materials: The GO colloidal suspension (~3 mg mL−1) was prepared by oxidizing graphite powder via a modified Hummers method.[1] Au-NPs with about 12 and 50 nm diameters, were prepared by sodium citrate reduction.[2-3] Zn2SnO4-NWs were prepared as our previously reported.[2-4] TiO2-NPs (P25) was purchased from Degussa. PANI-NFs were prepared as previously reported (HClO4 was used as dopant).[5] CNTs were purchased from CNano Technology Ltd. MnO2-NWs were prepared as previously reported.[6]

The fabrication of InN-NWs: InN-NWs were fabricated by in situ nitriding of In2O3 powder in a conventional furnace with a quartz tube mounted horizontally inside. In brief, full-grinding In2O3 powder was served as the starting material and was placed uniformly in a quartz boat. The boat was then loaded into the long quartz tube. The quartz tube was degassed and then purged with high-purity (99.999%) nitrogen. During the growth process, 100 sccm of NH3 (anhydrous, 99.999%) was introduced into the tube. The quartz tube was heated to 680 °C and then maintained at this temperature for 8 h. After reaction, the quartz tube was...
cooled down under the protection of nitrogen. A fluffy brown product was formed on the quartz boat.

**Fabrication of rGO hybrid architectures (GHAs):** Au-NP, PANI-NF and MnO$_2$-NW aqueous suspensions were first centrifuged, respectively. Then, these resulting solids were dispersed into GO suspensions to obtain the mixed suspensions, respectively. The other nanomaterials were directly added into the GO suspensions to obtain the mixed suspensions, followed by ultrasonication. After obtaining stable mixed suspensions, the vials with suspensions were put into a Teflon (polytetrafluoroethylene) vessel (50 mL). The vessel was then sealed in a stainless steel autoclave and heated to 180 °C for 3 h. Followed by freeze-drying, 3D porous GHAs were obtained. The mass ratio of nanomaterials to rGO sheets (R$_m$(nanomaterials : rGO)) in their GHAs is: R$_m$(CNTs : rGO) = 2.5; R$_m$(InN : rGO) = 2.5; R$_m$(Zn$_2$SnO$_4$ : rGO) = 0.01, 2.5; R$_m$(TiO$_2$ : rGO) = 0.5, 1.25, 2.5, 5, 7.5, 10; R$_m$(PANI : rGO) = 0.5, 1.25, 2.5, 5, 7.5, 10, 12.5. Since the mass of Au-NPs is too small to be measured by balance, volume ratio of Au-NP suspension (before centrifugation) to GO suspension (R$_v$(Au : GO)) were used to characterize the contents of Au-NPs in rGO/Au-NP hybrid architecture. In rGO/Au-NP hybrid architecture, R$_v$(Au : GO) is 6. In rGO/TiO$_2$-NP/InN-NW hybrid architecture, R$_m$(TiO$_2$ : InN : rGO) = 1 : 1 : 1. In rGO/Au-NP/TiO$_2$-NP/PANI-NF hybrid architecture, R$_m$(TiO$_2$ : PANI : rGO) is 2.5 : 2.5 : 1 and R$_v$(Au : GO) is 6. The morphology and the microstructures of GHAs were characterized by FE-SEM (JSM-7600F) and transmission electron microscopy (TEM, JEOL JEM-2100F). The Raman spectra were recorded with a spectrophotometer (WITec alpha 300 R) with operating wavelength of 532 nm. The x-ray diffraction (XRD) patterns were recorded using a Shimadzu XRD-6000 X-ray diffractometer with Cu K$_\alpha$ irradiation.

**Photocurrent responses of rGO/TiO$_2$-NP hybrid architectures:** Indium tin oxide (ITO) glass was used as current collector. rGO/TiO$_2$-NP hybrid architecture was fixed on ITO and conductively connected by silver conducting paste. The area of ITO around rGO/TiO$_2$-NP
A hybrid architecture was covered by nonconductive adhesive, taking care that a part (0.5-1 cm) of ITO was not covered to be used as the electrical contact of the electrode with the conductive wire. The photocurrent measurements were carried out with a CHI 660D workstation and Xe lamp (HAL-320). The electrolyte was 0.5 M Na₂SO₄ aqueous solution. A three-electrode configuration was employed, as depicted in Figure 3a, consisting of rGO/TiO₂ architecture, Ag/AgCl and a platinum wire, serving as the working, reference and counter electrodes, respectively. The potential of the working electrode against the Ag/AgCl counter electrode is set at 0 V.

**Fabrication of supercapacitors based on rGO/PANI-NF architectures:** Pure rGO and rGO/PANI-NF hybrid architectures were cut into slices to be directly used as the both electrodes of supercapacitors. The masses of pure rGO and rGO/PANI-NF hybrid electrodes are controlled in a range of 1.5-2 mg. Pt slices were used as current collectors. Filter paper and H₂SO₄ (1 M) served as a separator and electrolyte, respectively.

CVs of the supercapacitors were performed by CHI 660D instrument (CHI Instruments). The galvanostatic charge-discharge of the supercapacitors was carried out on a supercapacitor test system (Solartron, 1470E)

The specific capacitance \( C_{sc} \) of two-electrode supercapacitor system at different scan rates was estimated from the CV curves using the equation:

\[
C_{sc} = \frac{\int I dV}{m s V} \quad (S1)
\]

where \( I \) is the current, \( s \) is the scan rate and \( m \) is the total mass of the active electrode material.

The specific capacitances \( C_{sc} \) of the supercapacitor devices were calculated from constant current charge/discharge curves using equation:

\[
C_{sc} = \frac{I}{(dV/dt)m} \quad (S2)
\]

where \( I \) is the discharge current, \( dV/dt \) represents the slope of the discharge curve and \( m \) is the total mass of the active electrode material on both electrodes.
Since both electrodes in a supercapacitor are in series, the total capacitance ($C_t$) of supercapacitor can be expressed as:

\[
\frac{1}{C_t} = \frac{1}{C_1} + \frac{1}{C_2} \quad (S3)
\]

\[
C_t = (m_1 + m_2)C_{sc} \quad (S4)
\]

\[
C_1 = m_1 C_{spe} \quad (S5)
\]

\[
C_2 = m_2 C_{spe} \quad (S6)
\]

where $C_1$ and $C_2$ are the capacitances of both electrodes, respectively; $m_1$ and $m_2$ are the mass of the active electrode material on both electrodes, respectively; $C_{spe}$ is the specific capacitance of rGO/PANI-NF architecture.

Therefore, the specific capacitance $C_{spe}$ of rGO/PANI-NF architecture can be expressed as:

\[
C_{spe} = \frac{(m_1 + m_2)^2C_{sc}}{m_1 m_2} \quad (S7)
\]
Figure S1. (a) AFM image of a GO sheet. SEM images of (b) Au-NPs, (c) TiO$_2$-NPs, (d) PANI-NFs, (e) CNTs, (f) MnO$_2$-NWs, (g) InN-NWs, and (h) Zn$_2$SnO$_4$-NWs.
Table S1 The shape, size, solubility and preparing methods of different typical nanomaterials.

<table>
<thead>
<tr>
<th>Materials</th>
<th>Shape</th>
<th>Size</th>
<th>Solubility in water</th>
<th>Preparing method</th>
</tr>
</thead>
<tbody>
<tr>
<td>Au</td>
<td>NP</td>
<td>D: ~ 13 nm</td>
<td>Good</td>
<td>Solution-based method</td>
</tr>
<tr>
<td>TiO₂</td>
<td>NP</td>
<td>D: ~ 21 nm</td>
<td>Medium</td>
<td>Solution-based method</td>
</tr>
<tr>
<td>PANI</td>
<td>NF</td>
<td>D: ~ 100 nm L: ~ 500 nm-1μm</td>
<td>Poor</td>
<td>Solution-based method</td>
</tr>
<tr>
<td>CNTs</td>
<td>NT</td>
<td>D: ~ 5-50 nm L: several micrometers</td>
<td>Medium</td>
<td>CVD</td>
</tr>
<tr>
<td>MnO₂</td>
<td>NW</td>
<td>D: ~ 5-20 nm L: several micrometers</td>
<td>Poor</td>
<td>Solution-based method</td>
</tr>
<tr>
<td>InN</td>
<td>NW</td>
<td>D: ~ 100 nm L: ~ 5-20 μm</td>
<td>Poor</td>
<td>CVD</td>
</tr>
<tr>
<td>Zn₂SnO₄</td>
<td>NW</td>
<td>D: tens to 200 nm L: tens of micrometers</td>
<td>Poor</td>
<td>CVD</td>
</tr>
</tbody>
</table>
Figure S2. Optical images of GO/TiO₂-NP suspensions and rGO/TiO₂-NP hybrid architectures with different Rₓm(TiO₂ : rGO).
**Figure S3.** Optical images of GO/PANI-NF suspensions and rGO/PANI-NF hybrid architectures with different $R_m$(PANI : rGO).
Figure S4. (a) The SEM image of TiO$_2$-NPs attached on rGO surface in rGO/TiO$_2$-NP hybrid architecture. (b) Raman spectra of TiO$_2$ (P25), GO, rGO architecture and rGO/TiO$_2$-NP hybrid architecture. (c) XRD patterns of TiO$_2$ (P25), rGO architecture and rGO/TiO$_2$-NP hybrid architecture. (d-g) The elemental mapping of C, O and Ti for TiO$_2$-NPs attached on rGO surface in rGO/TiO$_2$-NP hybrid architecture.
Figure S5. SEM and optical (inset) images of (a) pure rGO architecture and GHAs based on (b) PANI-NFs, (c) CNTs, (d) MnO$_2$-NWs and (e) InN-NWs. (f) SEM image of rGO/Au-NP/TiO$_2$-NP/PANI-NF hybrid architecture. Insets are the optical images of rGO/Au-NP/TiO$_2$-NP/PANI-NF hybrid architecture (left) and mixed rGO/Au-NP/TiO$_2$-NP/PANI-NF suspension (right). To distinguish different nanomaterials by SEM at a scale, Au-NPs with about 50 nm diameter were used in rGO/Au-NP/TiO$_2$-NP/PANI-NF architecture. In all the other cases, diameter of Au-NPs is about 13 nm.
Figure S6. (a) The interaction between Au-NP and GO sheet. (b) The interaction between Au-NP and rGO sheet.
Figure S7. SEM images of rGO/TiO$_2$-NP hybrid architectures with different R$_m$(TiO$_2$ : rGO):

(a) 1.25, (b) 2.5, (c) 5, (d) 7.5.
Figure S8. SEM images of rGO/PANI-NF hybrid architectures with different $R_{n}(PANI: rGO)$: (a) 1.25, (b) 2.5, (c) 7.5, (d) 10.
Figure S9. (a) CV and (b) charge/discharge curves of rGO/PANI-NF hybrid architectures under different $R_m$(PANI : rGO): scan rate: 10 mV s$^{-1}$, charge/discharge current: 1 A g$^{-1}$.


