Supporting information

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Capacitance calculation

If the capacitances of the two electrodes, i.e. positive and negative, can be expressed as \( C_p \) and \( C_n \), respectively, the overall capacitance \( (C_T) \) of the entire cell can be expressed as eqn (1):

\[
\frac{1}{C_T} = \frac{1}{C_p} + \frac{1}{C_n}
\]  

(1)

In a symmetrical supercapacitor, \( C_p = C_n = C_0 \), where \( C_0 \) represents the per-electrode capacitance and in this study represents the capacitance of one electrode. So the relationship between \( C_T \) and \( C_0 \) should be as eqn (2):

\[
C_0 = 2C_T = \frac{2It}{V}
\]  

(2)

where \( I, t \) and \( V \) are charged current, \( t \) is the discharge time, \( V \) is the voltage drop upon discharging (excluding IR drop).

As a result, the per-electrode areal capacitance \( (C_{A0}) \) and volumetric capacitance \( (C_{v0}) \) of the device is shown in eqn (3) and eqn (4):

\[
C_{A0} = \frac{C_0}{A_0} = \frac{2It}{A_0} = \frac{4It}{A_TV} = 4C_{A-cell}
\]  

(3)

\[
C_{v0} = \frac{C_0}{V_0} = \frac{2I(t - t_{OMC})}{V_0} = \frac{4I(t - t_{OMC})}{V_TV} = 4C_{v-cell}
\]  

(4)

Where \( A_0 \) is the surface area of in one electrode, which is approximately considered to be a cylinder. \( A_T \) represents the total mass the whole cell, in which \( A_T = 2A_0 \). \( C_{A-cell} \) is the areal capacity of the whole cell. \( V_0 \) is the volume of one electrode the total volume of the device.
\( v_t = 2v_0 \). \( C_{v-cell} \) is the volumetric capacitance of the whole device.

The corresponding volumetric energy and power density are calculated through eqn (5) and (6):

\[
E_v = \frac{1}{2} C_{v-cell} V^2 = \frac{1}{8} C_{v0} V^2
\]  

(5)

\[
P_v = \frac{E_v}{t}
\]  

(6)

**Figure S1.** Digital image of a vial containing CVD gr/OMC dispersions stabilized by P123 surfactants.
Figure S2. Pore size distribution of CVD gr/OMC below 2 nm calculated from the NLDFT model.

Figure S3. XRD pattern of CVD gr after Ni etching.
Figure S4. Absorbance at 660 nm measured for CVD gr/OMC dispersions as a function of sonication time.

Figure S5. The relationship between line resistance and mass loading of CVD gr/OMC.
Figure S6. Ultimate tensile strength of various carbon coated cotton threads.

Figure S7. XRD patterns of MnO$_2$ spheres.
Figure S8. Bulk heterojunction (BJH) pore distribution spectra of the CT-CVD gr/OMC-MnO$_2$ composite electrode.

Figure S9. The relationship between surface area (a)/pore volume (b) and mass loading of MnO$_2$ nanoparticles on CT-CVD gr/OMC electrode.
Figure S10. Coulombic efficiency of CT-CVD gr/OMC-MnO$_2$ supercapacitor as a function of cycling number at a current density of 13.76 mA cm$^{-2}$.

Figure S11. (a) Specific capacitance as a function of scan rate (inverse square root) for CT-CVD gr/OMC-MnO$_2$ electrode. (b) Specific (inverse) capacitance as a function of scan rate (square root) for CT-CVD gr/OMC-MnO$_2$ electrode.
Figure S12. Bar graph of mass normalized capacitance of MnO2 in various thread-like supercapacitors with Faradaic insertion capacity (red) and Faradaic pseudocapacitive charging (blue) derived from Trasatti’s method.

Figure S13. EIS spectra of CT-CVD gr/OMC supercapacitor before and after 3000 cycles.
Figure S14 (a,b). EIS data of CT-CVD gr/OMC and CT-CVD gr/OMC- MnO$_2$ composites and the equivalent circuit diagram used for the fitting.

Figure S15. SEM image of TiO$_2$ nanowires

Figure S16. (a) Highlights of the response speed under 350 nm illumination. (b)
Highlights of the recovery speed under 350 nm illumination.

Table S1. Structural characterization data for CVD gr, OMC and CVD gr/OMC

<table>
<thead>
<tr>
<th>Sample</th>
<th>BET-surface area (m² g⁻¹)</th>
<th>Mesopore volume (cm³ g⁻¹)</th>
<th>Micropore volume (cm³ g⁻¹)</th>
<th>Micro/mesoporosity</th>
</tr>
</thead>
<tbody>
<tr>
<td>CVD-gr</td>
<td>91</td>
<td>0.09</td>
<td>0</td>
<td>-</td>
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<tr>
<td>OMC</td>
<td>236</td>
<td>0.5</td>
<td>0.9</td>
<td>1.8</td>
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<tr>
<td>CVD-gr/OMC</td>
<td>328</td>
<td>0.4</td>
<td>1.1</td>
<td>2.8</td>
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</tbody>
</table>

Table S2. Comparison of electrochemical performance of yarn/fiber-based supercapacitors.

<table>
<thead>
<tr>
<th>Ref.</th>
<th>Materials</th>
<th>Electrolyte</th>
<th>Voltage window</th>
<th>Pᵥ mW cm⁻³</th>
<th>Ev mWh cm⁻³</th>
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</thead>
<tbody>
<tr>
<td>73</td>
<td>PPy/ MnO₂/rGO</td>
<td>PVA/ H₃PO₄</td>
<td>0-0.8</td>
<td>16</td>
<td>1.1</td>
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<tr>
<td>16</td>
<td>ZnO/ MnO₂</td>
<td>PVA/LiCl</td>
<td>0-0.8</td>
<td>2.4</td>
<td>0.04</td>
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<tr>
<td>74</td>
<td>TiO₂/ MnO₂</td>
<td>PVA/LiCl</td>
<td>0-0.8</td>
<td>230</td>
<td>0.3</td>
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<tr>
<td>63</td>
<td>MnO₂/Carbon fiber</td>
<td>PVA/H₃PO₄</td>
<td>0-0.8</td>
<td>400</td>
<td>0.22</td>
</tr>
<tr>
<td>75</td>
<td>PPy</td>
<td>PVA/ H₃PO₄</td>
<td>0-0.8</td>
<td>270</td>
<td>1</td>
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<td>15</td>
<td>PEDOT/CNT</td>
<td>PVA/ H₃PO₄</td>
<td>0-0.8</td>
<td>38000</td>
<td>1.1</td>
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<tr>
<td>76</td>
<td>WO₃₋ₓ/MoO₃₋ₓ</td>
<td>PVA/ H₃PO₄</td>
<td>0-1.9</td>
<td>730</td>
<td>1.9</td>
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<td>77</td>
<td>Nickel fiber/Co₃O₄ nanowire</td>
<td>PVA/KOH</td>
<td>0-1.5</td>
<td>1470</td>
<td>0.62</td>
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<tr>
<td>78</td>
<td>NiCo₂O₄</td>
<td>PVA/KOH</td>
<td>0-1</td>
<td>17000</td>
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<td>MnO₂/graphene/carbon fiber</td>
<td>PAAK/KCl</td>
<td>0-1.6</td>
<td>200</td>
<td>0.9</td>
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<tr>
<td>14</td>
<td>CuO/AuPd/ MnO₂</td>
<td>PVA/KOH</td>
<td>0-0.8</td>
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<td>This work</td>
<td>CVD gr/OMC- MnO₂ cotton thread</td>
<td>PVA-BMIMCl- Li₂SO₄</td>
<td>0-1.5</td>
<td>300</td>
<td>2.7</td>
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