Supplementary Information

Highly Stretchable Microsupercapacitor Arrays with Honeycomb Structures for Integrated Wearable Electronic Systems

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### Supplementary Table S1. Performance comparison of MSCs developed in this work and previous studies.

<table>
<thead>
<tr>
<th>Electrode material</th>
<th>Electrode thickness (μm)</th>
<th>Electrolyte</th>
<th>Volumetric capacitance</th>
<th>Capacitance retention</th>
<th>Time constant τ₀ (ms)</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>SWCNT</td>
<td>0.28</td>
<td>PVA-H₃PO₄</td>
<td>1 F cm⁻³ @ 100 V s⁻¹</td>
<td>54% @ 0.05–100 V s⁻¹</td>
<td>6.3</td>
<td>This work</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>63% @ 1–100 V s⁻¹</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>72% @ 1–50 V s⁻¹</td>
<td></td>
<td></td>
</tr>
<tr>
<td>RGO-CNT (9:1)</td>
<td>6</td>
<td>3 M KCl</td>
<td>3 F cm⁻³ @ 50 V s⁻¹</td>
<td>60% @ 1–50 V s⁻¹</td>
<td>4.8</td>
<td>[1]</td>
</tr>
<tr>
<td>Onion-like carbon</td>
<td>7</td>
<td>1 M Et₄NBF₄/anhydrous C₄H₆O₃</td>
<td>~0.7[ᵇ] F cm⁻³ @ 100 V s⁻¹</td>
<td>~53% @ 1–100 V s⁻¹</td>
<td>26</td>
<td>[2]</td>
</tr>
<tr>
<td>SWCNT</td>
<td>~3.5</td>
<td>3.6 M H₂SO₄</td>
<td>1.04 F cm⁻³</td>
<td></td>
<td>7</td>
<td>[3]</td>
</tr>
<tr>
<td>Direct laser-write porous carbon</td>
<td>33[ᵃ]</td>
<td>PVA-H₃PO₄</td>
<td>0.24 F cm⁻³ @ 0.01 V s⁻¹</td>
<td>~3%[ᵇ] @ 0.01–100 V s⁻¹</td>
<td></td>
<td>[4]</td>
</tr>
<tr>
<td>Nanoporous carbon</td>
<td>1–40</td>
<td>0.2 M K₂SO₄</td>
<td>2.0[ᶜ] F cm⁻³ @ 1 V s⁻¹</td>
<td>15% @ 0.01–1 V s⁻¹</td>
<td></td>
<td>[5]</td>
</tr>
<tr>
<td>RGO</td>
<td>0.025</td>
<td>PVA-H₃PO₄</td>
<td>359 F cm⁻³ @ 0.005 V s⁻¹</td>
<td>~42%[ᵇ] @ 0.005–1 V s⁻¹</td>
<td></td>
<td>[6]</td>
</tr>
<tr>
<td>Graphene microribbon</td>
<td>0.1–0.44</td>
<td>PVA-H₂SO₄</td>
<td>(13–20)[ᶜ] F cm⁻³ @ 0.5 V s⁻¹</td>
<td>~40%[ᵇ] @ 0.05–1 V s⁻¹</td>
<td></td>
<td>[7]</td>
</tr>
<tr>
<td>ERGO</td>
<td>20</td>
<td>25% KOH</td>
<td>0.24 F cm⁻³ @ 40 μA cm⁻²</td>
<td>~93% @ 40–800 μA cm⁻²</td>
<td>1.35</td>
<td>[8]</td>
</tr>
<tr>
<td>va-MWCNT</td>
<td>15</td>
<td>LiPF₆</td>
<td>~0.11[ᵈ] F cm⁻³ @ 100 V s⁻¹</td>
<td>~41%[ᵈ] @ 1–100 V s⁻¹</td>
<td>2.23</td>
<td>[9]</td>
</tr>
</tbody>
</table>

RGO = reduced graphene oxide; ERGO = electrochemically reduced graphene oxide; va-MWCNT = vertically aligned multi-walled carbon nanotubes

[a] Estimated from volumetric and areal capacitance data from the literature.
[b] Estimated from specific capacitance vs scan rate data from the literature.
[c] Estimated from areal capacitance and electrode thickness data from the literature.
[d] Estimated from the CV curves and electrode thickness.
Supplementary Figure S1. Distribution of maximum (first principal) strain $\varepsilon_{\text{max}}$ in a four-cell honeycomb PDMS substrate stretched by 100%.
Supplementary Figure S2. (a) Schematic diagram showing the location of MSCs in a $4 \times 4$ MSC array and (b) corresponding circuit diagram.
**Supplementary Figure S3.** Fabrication process of stretchable $4 \times 4$ MSC arrays: (a) spin coating of the bottom PI layer on a Si wafer; (b) evaporation and micropatterning of the Cr/Au bilayer; (c) spin coating and micropatterning of the top PI layer; (d) spray deposition and micropatterning of the interdigital SWCNT electrodes; (e) selective etching of the bottom PI layer; (f) transfer of the device onto a honeycomb PDMS substrate; (g) coating of the interdigital SWCNT electrodes with gel electrolyte; and (h) application of the top honeycomb PDMS superstrate to encapsulate the MSC array.
**Supplementary Figure S4.** Digital photographs of a stretchable 4 × 4 MSC array at characteristic fabrication stages: (a) patterned Cr/Au bilayer on a PI-coated Si substrate; (b) patterned interdigital SWCNT electrodes on top of the interdigital Cr/Au bilayer pattern; (c) peeled off device attached to a water dissolvable tape; and (d) device transferred onto the honeycomb bottom PDMS substrate.
Supplementary Figure S5. Digital photograph of an undeformed 3 × 3 MSC array.
Supplementary Figure S6. (a) Raman and (b) FTIR spectra of SWCNTs. For the Raman spectrum shown in (a), $I_D/I_G = 0.078$. The peaks at 1112, 1635, and 3448 cm$^{-1}$ in the FTIR spectrum shown in (b) are assigned to $–$CH bending,$^{10,11}$ CNT back bone,$^{12}$ and carboxylate $–$OH stretching,$^{11}$ respectively.
Supplementary Figure S7. CV curves of Au and Au + SWCNT single MSCs for a scan rate of (a) 10, (b) 50, and (c) 100 V s\(^{-1}\) and (d) volumetric capacitance \(C_v\) versus scan rate of SWCNT and Au + SWCNT single MSCs.
Supplementary Figure S8. CV curves of a 3 × 3 MSC array for a scan rate of 10, 50, and 100 V s⁻¹.
Supplementary Figure S9. CV curves of a 3 × 3 MSC array obtained soon after fabrication (day 0) and 5 days later.
Supplementary Figure S10. Schematic illustration of regions referred to as the islands and the beams of a 4 × 4 MSC array.
Supplementary Figure S11. Distribution of maximum (first principal) strain $\varepsilon_{\text{max}}$ in the (a) bottom PDMS, (b) bottom PI, (c) Au, and (d) top PI layers of a 4 × 4 MSC array stretched by 150%.
Supplementary Figure S12. Optical images of a $3 \times 3$ MSC array at different deformation stages (left column) and corresponding FEA results of maximum (first principal) strain $\varepsilon_{\text{max}}$ in the Au layer (right column).
Supplementary Figure S13. Distribution of maximum (first principal) strain $\varepsilon_{\text{max}}$ in the (a) bottom PDMS, (b) bottom PI, (c) Au, and (d) top PI layers of a 3 × 3 MSC array stretched by 100%.
Supplementary Figure S14. Capacitance retention versus elongation of a 3 × 3 MSC array (the inset figure shows CV curves for −50% compression and 0–100% elongation).
Supplementary Figure S15. FEA simulations of a three-cycle stress-strain response of the (a) PI layer, (b) Au capacitor, and (c) Au interconnect of a 4 × 4 MSC array stretched up to 100%.
Supplementary Figure S16. A $3 \times 3$ MSC array integrated with a power management chip on a honeycomb substrate.
Supplementary Figure S17. Digital photographs showing tensile test setups of (a) a single honeycomb cell of the device, (b) flat PDMS membrane, and (c) cloth strip cut from a Nike wrist band.
**Supplementary Figure S18.** Distribution of maximum (first principal) strain $\varepsilon_{\text{max}}$ in the PDMS layer of a $4 \times 4$ MSC array (a) bent by $180^\circ$ and (b) twisted by $60^\circ$. 
Supplementary Figure S19. Maximum (first principal) strain $\varepsilon_{\text{max}}$ in the Au layer of a $4 \times 4$ MSC array as a function of (a) bending and (b) twisting angle. The inset figures in (a) show side-view schematics of the deformed device for $0^\circ$, $90^\circ$, and $180^\circ$ bending angle, whereas the inset figures in (b) show schematics of the deformed device for $0^\circ$, $30^\circ$, and $60^\circ$ twisting angle.
Supplementary References


