Supporting Information

A Multidimensional Nanostructural Design towards Electrochemically Stable and Mechanically Strong Hydrogel Electrodes

Wei Zhang, †* Jing Ma,† Wenjuan Zhang, Peigen Zhang, Wei He, Jian Chen, ZhengMing Sun*

† Prof. W. Zhang, J. Ma, Prof. P. G. Zhang, Prof. W. He, Prof. Z. M. Sun
† Jiangsu Key Laboratory of Advanced Metallic Materials
† School of Materials Science and Engineering
Southeast University, Nanjing, 211189, PR China
E-mail: w69zhang@seu.edu.cn; zmsun@seu.edu.cn
Prof. W. J. Zhang
State Key Laboratory for Advanced Processing and Recycling of Non-ferrous Metals
Lanzhou University of Technology, Lanzhou, 730050, PR China
Part 1. Calculation

Mechanical Test

The stress (\(\sigma\)) (MPa), strain (\(\varepsilon\)) (100%), elastic modulus (\(E\)) (MPa) and deformation energy (\(W\)) (MJ m\(^{-3}\)) of all MXene-based hydrogels were calculated according to eqs 1, 2, 3 and 4:

\[
\sigma = \frac{P}{A} \quad (1)
\]
\[
\varepsilon = \frac{\Delta L}{L} \times 100 \quad (2)
\]
\[
E = \frac{\sigma}{\varepsilon} \quad (3)
\]
\[
W = \int_0^\varepsilon \sigma d\varepsilon \quad (4)
\]

where \(P\) (N) is the maximum load along the direction of applied force at fracture, \(A\) (mm\(^2\)) is the cross-sectional area of the fracture, \(\Delta L\) is breaking elongation and \(L\) is the original length.

Electrochemical Characterization

The electrochemical performances of all MXene-based hydrogel electrodes were performed in a three-electrode system. The work electrode was MXene-based hydrogels (size of 10 mm \(\times\) 10 mm \(\times\) 2 mm), the reference electrode was Hg/Hg\(_2\)SO\(_4\) electrode (Shanghai Lei Magnetism Instrument Co., Ltd.), and the counter electrode was a titanium plate, respectively.

The specific capacitance (\(C_p\)) (F.g\(^{-1}\)) of all electrodes were calculated according to their GCD curves and derived from eq 5:

\[
C_p = \frac{I \times \Delta t}{m \times \Delta V} \quad (5)
\]

where \(I\) is the discharge current, \(\Delta t\) is the discharge time of CGD curves, \(m\) is the mass of active materials in single working electrodes, and \(\Delta V\) is the voltage change during discharge.

For flexible symmetric solid-state supercapacitors, the electrochemical performance were measured in a two electrode system. The cell-specific capacitance (\(C_{cell}\)) (F.g\(^{-1}\)) of all solid-state supercapacitor devices were calculated from their CGD curves according to eq 6:

\[
C_{cell} = \frac{I \times \Delta t}{M \times \Delta V} \quad (6)
\]

where \(I\) is the discharge current, \(\Delta t\) is the discharge time of CGD curves, \(M\) is the mass of active materials in two pieces working electrodes, and \(\Delta V\) is the voltage change during discharge.

The energy density (\(E_{cell}\)) and power density (\(P_{cell}\)) of the supercapacitor devices were calculated based on eqs 7 and 8:

\[
E_{cell} = \frac{C_{cell} \times (\Delta V)^2}{2 \times 3.6} \quad (7)
\]
\[ P_{\text{cell}} = \frac{E_{\text{cell}} \times 3600}{\Delta t} \quad (8) \]

where \( E_{\text{cell}} \) is the energy density, \( P_{\text{cell}} \) is the power density and \( C_{\text{cell}} \) is the cell-specific capacitance, the \( \Delta V \) is the voltage change during discharge, the \( \Delta t \) is the discharge time from GCD curves.

**Part 2. Figures**

**Figure S1:** SEM images of MXene-PVA hydrogels with different MXene concentrations: (a) 0.2 mg cm\(^{-3}\); (b) 0.4 mg cm\(^{-3}\); and (c) 2.0 mg cm\(^{-3}\).

**Figure S2.** (a) XRD patterns of the PVA, PPy, MXene-PVA, MXene/PPy-PVA and MXene; (b) FTIR spectrum of the MXene/PPy-PVA hydrogel.
Figure S3. (a) The Maximum tensile stress of MXene-PVA hydrogels with different MXene concentrations, (b) Compression stress-strain curves of PVA and different concentration MXene-PVA hydrogels.

Figure S4. Electrochemical characterizations of 0.2 mg cm\(^{-3}\) MXene hydrogel electrodes: (a) CV, (b) charge/ discharge, and (c) EIS curves.

Figure S5. Electrochemical characterizations of 0.4 mg cm\(^{-3}\) MXene hydrogel electrodes: (a) CV, (b) charge/ discharge, and (c) EIS curves.
Figure S6. Electrochemical characterizations of 2 mg cm$^{-3}$ MXene hydrogel electrodes: (a) CV, (b) charge/discharge, and (c) EIS curves.

Figure S7. Specific capacitance and volume capacitance of MXene-PVA electrodes with different MXene concentration.
Figure S8. Electrochemical characterizations of MXene/PPy-PVA hydrogel electrodes: (a) CV, (b) charge/discharge, (c) EIS curves, and (d) The cycle life and coulombic efficiency.

Figure S9. EIS curves of the assembled flexible supercapacitor.
Figure S10. Photographs of PVA and MXene/PPy-PVA hydrogels during tensile tests: (a), (b) and (c) were PVA hydrogels; (d), (e) and (f) were MXene/PPy-PVA hydrogels.

Part 3. Tables

Table S1. Mass specific capacitance and tensile strength of different hydrogel electrode materials

<table>
<thead>
<tr>
<th>Hydrogel electrodes</th>
<th>Specific capacitance (F g⁻¹)</th>
<th>Tensile strength (MPa)</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>PANI@CNF-PVA</td>
<td>201.6 at 1 A g⁻¹</td>
<td>0.032</td>
<td>1</td>
</tr>
<tr>
<td>PVAB@CNT-CN</td>
<td>117.1 at 1 A g⁻¹</td>
<td>0.093</td>
<td>2</td>
</tr>
<tr>
<td>PANI</td>
<td>750 at 1 A g⁻¹</td>
<td>0.600</td>
<td>3</td>
</tr>
<tr>
<td>Polythiophene</td>
<td>135 at 1 A g⁻¹</td>
<td>160</td>
<td>4</td>
</tr>
<tr>
<td>PANI@CNTs@PLA</td>
<td>510.3 at 1 A g⁻¹</td>
<td>18.7</td>
<td>5</td>
</tr>
<tr>
<td>CTS@SA</td>
<td>234.6 at 1 A g⁻¹</td>
<td>0.290</td>
<td>6</td>
</tr>
<tr>
<td>PANI@GO</td>
<td>115.2 at 1 A g⁻¹</td>
<td>351.9</td>
<td>7</td>
</tr>
<tr>
<td>Graphene</td>
<td>175 at 1 A g⁻¹</td>
<td>0.450</td>
<td>8</td>
</tr>
<tr>
<td>MXene@PPy-PVA</td>
<td>614 at 1 A g⁻¹</td>
<td>10.3</td>
<td>our work</td>
</tr>
</tbody>
</table>


Part 4. Movies

For the movies, Movie S1 and S2 show the stretching properties of pure PVA and MXene-PVA hydrogel cylinders. The pure PVA hydrogel can be broken easily, while the MXene-PVA shows strong mechanical strength. Movie S3 and S4 show the cyclic compression of pure PVA and MXene-PVA hydrogel cylinders. The pure PVA hydrogel can be broken easily, while the MXene-PVA shows strong compression strength.
References