Supporting Information for

Intercalation and Surface Modification of Two-dimensional Transition Metal Carbonitride Ti$_3$CNT$_x$ for Ultrafast Supercapacitors

Shuaikai Xu*, Zhemin Li*, Guodong Wei, Yuanhao Wang*, Ya Yang*ab

*aGuangxi Novel Battery Materials Research Center of Engineering Technology, Guangxi Colleges and Universities Key Laboratory of Novel Energy Materials and Related Technology, Center on Nanoenergy Research, College of Physics Science and Technology, Guangxi University, Nanning 530004, P. R. China. E-mail: skxu@gxu.edu.cn

bCAS Center for Excellence in Nanoscience, Beijing Key Laboratory of Micro-nano Energy and Sensor, Beijing Institute of Nanoenergy and Nanosystems, Chinese Academy of Sciences, Beijing 101400, P. R. China. E-mail: yayang@binn.cas.cn

cSUSTech Engineering Innovation Center, School of Environmental Science and Engineering, Southern University of Science and Technology, Shenzhen, Guangdong 518055, P. R. China. E-mail: wangyh2020@mail.sustech.edu.cn

dMaterials Institute of Atomic and Molecular Science, School of Physics and Energy, Shaanxi University of Science and Technology, Xi’an 710021, Shaanxi, People’s Republic of China.
**Supplementary Table:**

**Table S1** BET specific surface area and pore size of the Ti₃CNTₓ-based films

<table>
<thead>
<tr>
<th></th>
<th>Ti₃CNTₓ</th>
<th>300-Ti₃CNTₓ</th>
<th>K-Ti₃CNTₓ</th>
<th>300-K-Ti₃CNTₓ</th>
<th>300-K-Ag-Ti₃CNTₓ</th>
</tr>
</thead>
<tbody>
<tr>
<td>Specific surface area (m² g⁻¹)</td>
<td>63.18</td>
<td>75.44</td>
<td>70.85</td>
<td>90.37</td>
<td>82.63</td>
</tr>
<tr>
<td>Pore size (nm)</td>
<td>3.54</td>
<td>3.62</td>
<td>3.86</td>
<td>3.73</td>
<td>3.81</td>
</tr>
</tbody>
</table>

**Table S2** Simulation results of the EIS spectra in Figure 4f

<table>
<thead>
<tr>
<th></th>
<th>Ti₃CNTₓ</th>
<th>300-Ti₃CNTₓ</th>
<th>K-Ti₃CNTₓ</th>
<th>300-K-Ti₃CNTₓ</th>
<th>300-K-Ag-Ti₃CNTₓ</th>
</tr>
</thead>
<tbody>
<tr>
<td>$R_1$/Ohm</td>
<td>4.2</td>
<td>5.1</td>
<td>2.9</td>
<td>3.2</td>
<td>3.0</td>
</tr>
<tr>
<td>$R_2$/Ohm</td>
<td>0.3</td>
<td>0.6</td>
<td>6.1</td>
<td>9.7</td>
<td>0.2</td>
</tr>
</tbody>
</table>
Supplementary Figures:

**Fig. S1.** (a) Nitrogen adsorption-desorption isotherms and (b) pore size distributions of the Ti$_3$CNT$_x$-based films.

**Fig. S2.** (a) CVs at 20 mV$^{-1}$ and (b) rate performance of the 300-K-Ag-Ti$_3$CNT with different AgNWs contents.
**Fig. S3.** (a) Comparison of the cycling performance of the Ti$_3$CNT$_x$-based films prepared in this work. (b) The equivalent circuit used for fitting impedance spectra of the Ti$_3$CNT$_x$-based films. $R_1$: equivalent series resistance; $C$: electrical capacitor; $R_2$: charge transfer resistance; CPE: constant phase angle element, and $Z_W$: Warburg impedance.

**Fig. S4.** (a) The CV curves at 20 mV s$^{-1}$ and (b) digital photographs of the flexible all-solid-state supercapacitors under different bending angles.
Calculation method of capacitive contribution:

The response current at a fixed potential can be expressed as being the combination of surface capacitive effects and diffusion-controlled insertion processes:

\[ i(V) = k_1 V + k_2 V^{1/2} \]

We rearrange this slightly to:

\[ \frac{i(V)}{V^{1/2}} = k_1 V^{1/2} + k_2 \]

\( k_1 V \) and \( k_2 V^{1/2} \) represent the current contributions from the surface capacitive effects and the diffusion-controlled intercalation process, respectively. Determining \( k_1 \) and \( k_2 \), the fraction of the current due to each of these contributions can be quantified. The detailed procedures to evaluate the \( C_{\text{capacitive}} \) and \( C_{\text{diffusion}} \) are as follows: (1) The CVs at very slow scan rates (<20 mV s\(^{-1}\)) should be collected; (2) Fix a potential and read the current from different CVs; (3) Plot the lines \( \frac{i(V)}{V^{1/2}} \) vs. \( V^{1/2} \). The \( k_1 \) and \( k_2 \) are the slope and y-intercept, respectively; (4) Differentiate current at a fixed scan rate; (5) Repeat steps (3-4) for other potentials; (6) Calculate \( C_{\text{capacitive}} \) and \( C_{\text{diffusion}} \).