Experimental supplementary information

Electrophoresis-Microwave Synthesis of S, N-Doped Graphene Foam for High-Performance Supercapacitors

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Experimental section

Characterization

The specific surface area of the samples was measured by methylene blue (MB) adsorption method with UV-vis spectroscopy: The standard curve of absorbance versus MB concentration was obtained by measuring 10 parts of MB standard solution with gradient concentration at the wavelength of 658 nm. The dGFs with gradient mass were ultrasonically dispersed into MB solution with known concentration. After stirring at room temperature for 24 hours, the supernatants were centrifuged to obtain MB solutions with adsorption equilibrium. Measure the absorbance of the obtained MB solutions at 658 nm, and calculate its concentration according to the standard curve. According to Langmuir adsorption equation (1):

\[
\Gamma = \Gamma_\infty \frac{KC}{1+KC}, \quad \frac{1}{\Gamma} = \frac{1}{\Gamma_\infty} + \frac{1}{KC_\infty} \cdot \frac{1}{C} \quad (1)
\]

Where \( \Gamma \) is the mass of MB adsorbed per unit mass of dGF under adsorption equilibrium (g g\(^{-1}\)), \( \Gamma_\infty \) is the mass of MB adsorbed by unit mass DGF under saturated adsorption (g g\(^{-1}\)), \( K \) is the adsorption coefficient, \( C \) is the concentration of MB at adsorption equilibrium (g L\(^{-1}\)). Draw and fit the relationship between \( 1/\Gamma \) and \( 1/C \) to get the value of \( \Gamma_\infty \) and \( K \). Due to the coverage of MB molecular on the surface of \( sp^2 \) hybrid carbon layer is 2450 m\(^2\) g\(^{-1}\), the specific surface area (SSA, m\(^2\) g\(^{-1}\)) of dGFs can be calculated by the formula (2):

\[
SSA = 2450\Gamma_\infty \quad (2)
\]

Electrochemical measurements

The specific capacitance of the electrode material was calculated from discharge curves according to the following equation (Eq.(3)):

\[
C_m = \frac{I\Delta t}{m\Delta V} \quad (3)
\]
where \( C_m \) is the specific capacitance (F g\(^{-1}\)), \( I \) is the constant discharge current (A), \( \Delta t \) is the discharge time (s), \( m \) is the total mass of the active material (g) and \( \Delta V \) is the set potential window (V). The energy density (\( E_m \)) and power density (\( P_m \)) were calculated according to the following equation (Eq.(4) and Eq.(5)):

\[
E_m = \frac{C_m U^2}{7.2} \quad (4)
\]

\[
P_m = \frac{3.6E_m}{\Delta t} \quad (5)
\]

**Results and Discussion**

![Figure S1](image1.png)

**Figure S1.** (a) High-resolution FE-SEM image and (b) EDS of PSS-GO.

![Figure S2](image2.png)

**Figure S2.** FT-IR spectra of GO and PSS-GO.
Figure S3. LSV curve of the electrolyte for EPD.

Figure S4. Photos of EPD cells in the first 1 min at voltage of 1.2 V and 3.5 V.

Figure S5. Digital photo of GOG.
**Figure S6.** Digital photos of the GOF and the dGF.

**Figure S7.** FE-SEM images of GOF and dGF.

**Figure S8.** High-resolution FE-SEM image of dGF.
Figure S9. Full XPS survey of (a) GOF and (b) dGF.

Figure S10. (a) C 1s, (b) N 1s, and (c) S 2p XPS survey of GOF.

Figure S11. (a) Deposition current-time curves under different EPD voltages.
**Figure S12.** (a-g) Cross-sectional FE-SEM images of GOF prepared under different EPD voltages.

**Figure S13.** The relationship of deposition thickness and time under different EPD voltages.
Figure S14. Superficial FE-SEM image of the GOF-1.0-4.0.

Figure S15. XRD patterns of GOF with different EPD voltages.

Figure S16. The nitrogen adsorption/desorption isotherm and pore distribution for dGF-1.0-3.5.
Figure S17. CV curves and GCD curves of dGFs with different EPD voltages.
Figure S18. Randles equivalent circuit representing the circuit elements in the EIS.

Figure S19. The fitting result of CV log (current density) to log (sweep rate) of dGFs with different EPD voltages.

Figure S20. The contact angle measuring images of dGF with different PSS dosage.
Figure S21. CV curves and GCD curves of dGFs with different PSS dosages.
Figure S22. The CV curves of dGFs with different PSS dosage under 50 mV s⁻¹.

Figure S23. The fitting result of CV log (current density) to log (sweep rate) of dGFs with different EPD voltages.

Figure S24. The preparation process of the interdigital electrodes.

1 Electrophoretic deposition

2 Microwave treatment
**Figure S25.** GCD curves at 1 A g\(^{-1}\) of the assembled interdigital supercapacitor under different bending angles.

**Figure S26.** Photos of the assembled interdigital supercapacitor lighting up the LED bulb under (a-e) different bending angles and (f) twisting state.

**Figure S27.** EIS of the assembled interdigital supercapacitor before and after 10000 cycles.
**Figure S28.** The shape of interdigital electrodes.

<table>
<thead>
<tr>
<th>dGFs with different EPD voltage</th>
<th>Equivalent series resistance, $R_S$ (Ω)</th>
<th>Charge transfer resistance, $R_{CT}$ (Ω)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$d$GF-$1.0$-$2.0$</td>
<td>0.23</td>
<td>6.16</td>
</tr>
<tr>
<td>$d$GF-$1.0$-$3.0$</td>
<td>0.34</td>
<td>7.05</td>
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<tr>
<td>$d$GF-$1.0$-$3.5$</td>
<td>0.41</td>
<td>6.19</td>
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<tr>
<td>$d$GF-$1.0$-$4.0$</td>
<td>0.32</td>
<td>8.99</td>
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**Table S1.** The EIS fitting results of $d$GFs with different EPD voltage.

<table>
<thead>
<tr>
<th>dGFs with different PSS dosage</th>
<th>Equivalent series resistance, $R_S$ (Ω)</th>
<th>Charge transfer resistance, $R_{CT}$ (Ω)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$d$GF-$0.4$-$3.5$</td>
<td>0.28</td>
<td>5.76</td>
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<td>$d$GF-$0.6$-$3.5$</td>
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<td>5.17</td>
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<td>$d$GF-$0.8$-$3.5$</td>
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<td>6.32</td>
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<td>$d$GF-$1.0$-$3.5$</td>
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<td>$d$GF-$1.2$-$3.5$</td>
<td>0.31</td>
<td>5.66</td>
</tr>
</tbody>
</table>

**Table S2.** The EIS fitting results of $d$GFs with different PSS dosage.