Supporting Information

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Bioinspired Layered Composites Based on Flattened Double-Walled Carbon Nanotubes

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Supporting Information

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**Figure S1.** (a) Set-up for synthesizing FDWCNT films; (b) The size of a FDWCNT film is 16 cm (length) × 5 cm (width), and the thickness can be tuned with the synthesizing time. In this work, the thickness of the FDWCNT film is about 500 µm, and the synthesis time is controlled to be 30 min; (c) The FDWCNT film can be folded into any shape such as a small airplane without any damage, indicating that it is very tough and flexible.
Figure S2. Raman spectrum of FDWCNTs indicates four radial breathing mode (RBM) peaks at 133, 188, 249, and 279 cm$^{-1}$, respectively, confirming the existence of double walled carbon nanotubes.[S1] The peaks at 1322 cm$^{-1}$ and 1590 cm$^{-1}$ are assigned to the disorder-induced D-band and tangential mode G-band, respectively. The intensity ratio of D-band to G-band ($I_D/I_G$) is only 0.11, indicating that the obtained FDWNTs are of high quality and have well-ordered structure.[S1]
Figure S3. Thermogravimetric analysis (TGA) in air: (a) As-synthesized FDWCNTs with the purity of 82 wt%; (b) After purification, the purity of FDWCNTs can reach as high as 95 wt%.
Figure S4. (a) G-band peaks of FDWCNTs after stretching at different detection angles, in respect to the direction of laser polarization. (b) G-band intensity as a function of the detection angles, which are normalized to the intensity at $0^\circ$. 
Figure S5. Comparison of FT-IR spectra of (a) FDWCNTs, (b) epoxidation-functionalized FDWCNTs, and (c) epoxidation-functionalized FDWCNT/epoxy composites. The peak at 1210 cm\(^{-1}\) is attributed to epoxide ring groups, confirming that the epoxide groups are successfully grafted onto the surface of FDWCNTs. After reaction with epoxy resin in the curing process, the peak at 1210 cm\(^{-1}\) disappears, indicating occurrence of the cross-linking reaction between epoxidation-functionalized FDWCNTs and epoxy resin.
Figure S6. TGA comparison of (a) FDWCNTs, (b) aligned FDWCNT/epoxy composites with cross-linking, and (c) epoxy resin. Gas: nitrogen.
Figure S7. Comparison of different packing ways between (a) the cylindrical DWCNTs and (b) the flattened DWCNTs (e.g., FDWCNTs). It is obvious that FDWCNTs occupy more volume fractions compared to the cylindrical DWCNTs.
Figure S8. Typical stress-strain curves of pure FDWCNT films: (a) The tensile strength, Young’s modulus and strain of random FDWCNTs film are 135 ± 15 MPa, 0.70 ± 0.3 GPa and 25 ± 4 %, respectively. (b) After alignment by stretching, the tensile strength and Young’s modulus increase to 300 ± 25 MPa and 6.20 ± 1.2 GPa, respectively, while the strain decreases to 7.5 %.
Step 1: The FDWCNT is functionalized with epoxide groups by peroxide acid m-CPBA. Step 2: The epoxide groups on the FDWCNT are firstly attacked by accelerator BDMA, yielding zwitterions that contain a quaternary nitrogen cation and an active anion. Step 3: The anion

Figure S9. Proposed cross-linking mechanism between FDWCNT and epoxy resin system.
attacks the curing agent MTHPA, leading to formation of a species bearing a carboxylate anion as the active site. Step 4: This type of ester is functionalized as the initiator of the chain-wise polymerization, and then reacts with epoxy resin GE, leading to formation of an alkoxide species. Subsequently, the formed alkoxide species attacks the curing agent of MTHPA again. It should be stressed that the reaction in step 4 can happen at both ends of GE with the functionalized FDWCNT, eventually resulting in formation of FDWCNT-epoxy composite in chemical cross-linking bonds.\textsuperscript{[S2]}

Table S1. Mechanical properties of epoxy resin matrix

<table>
<thead>
<tr>
<th>Properties</th>
<th>Epoxy resin</th>
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<tbody>
<tr>
<td>Tensile strength (MPa)</td>
<td>83 ± 5</td>
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<tr>
<td>Young’s modulus (GPa)</td>
<td>2.2 ± 0.3</td>
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<tr>
<td>Strain (%)</td>
<td>7.88 ± 0.14</td>
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<tr>
<td>Toughness (MJ/m\textsuperscript{3})</td>
<td>4.6 ± 0.1</td>
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</tbody>
</table>

References
