Super-resilient and highly sensitive graphene oxide/cellulose-derived carbon aerogel

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S1. Details on experimental methods

**Materials.** Hydroxypropyl methyl cellulose (HPMC) was purchased from Macklin, China. Graphite powders were purchased from Nanjing XFNANO MaterialsTech Co., Ltd. 98 wt% H$_2$SO$_4$ and KMnO$_4$ were purchased from Aladdin, China.

**Preparation of HPMC/GO suspension.** GO was prepared through oxidizing graphite powders via a modified Hummers method$^1$, and then the obtained GO suspension was ultrasonicated (KQ-100B) for 10 min. To obtain HPMC/GO suspension, 1.08 g HPMC was dissolved in 33.92 mL deionized water, and then added 10.00 g GO suspension (1.8 wt%, 0.18 g GO). The above mixture was stirred for 12 h (500-600 rpm) and then ultrasonicated for another 1 h to make sure the remove of air bubbles and the homogeneous mixing of HPMC and GO.

**Fabrication of aerogels.** HPMC/GO suspension (15 mL) was poured into a plastic box (38 mm × 29 mm × 22 mm), and then the box was placed on a steel box filled with liquid nitrogen. After directional freeze-casting and freeze-drying (in a lyophilizer for 48 h), an aerogel (A-HPMC/GO-6, A and 6 represent aerogel and the mass ratio of HPMC to GO, respectively) was obtained. A-HPMC/GO-x with same solid content but different mass ratios of HPMC to GO (x= 1, 2, 4, 6, 8, 10 and 20, representing 1:1, 2:1, 4:1, 6:1, 8:1, 10:1 and 20:1, respectively) were prepared by the same method. In addition, pure HPMC aerogel (A-HPMC) and pure GO aerogel (A-GO) were fabricated from pure HPMC solution and GO suspension. To investigate the effect of directional freezing-casting on the structure and mechanical performance of carbon aerogel, control sample A-R-HPMC/GO-6 was prepared via a random freeze-drying method by immersing a plastic box containing HPMC/GO suspension in liquid nitrogen and then freeze-drying.

**Pre-oxidation of aerogels.** The aerogel was heated to 240 °C$^2$, 3 (0.5 °C min$^{-1}$) in air using a muffle furnace and held for 4 h to obtain pre-oxidized aerogels before carbonization.

**Fabrication of carbon aerogels.** The pre-oxidized aerogel was carbonized in a tube furnace under N$_2$ atmosphere. There are three stages during carbonization. The first stage was carried out from room temperature to 300 °C (5 °C min$^{-1}$). In the second stage, the sample was pyrolyzed from 300 °C to 400 °C at a heating rate of v °C min$^{-1}$ (v = 0.5, 1, 3, respectively) and held at 400 °C for 1 h. In the last stage, aerogel was heated to 750 °C (5 °C min$^{-1}$) and kept at 750 °C for 2 h to obtain a carbon aerogel. Aerogels (A-HPMC/GO-x, A-HPMC, A-GO and A-R-HPMC/GO-6) treated by both pre-oxidation and carbonization were named as C-HPMC/rGO-x, C-HPMC, C-rGO and C-R-HPMC/rGO-6, respectively. For comparison, C-N-HPMC/rGO-6 was prepared by directly carbonizing A-HPMC/GO-6 (without pre-oxidation).

**Characterizations.** The morphologies of all aerogels were observed on scanning electron microscopy (SEM, Merlin, Zeiss) and transmission electron microscopy (TEM, JEM-2100F). Thermal gravity analysis (TGA) was performed on a Pyris Diamond TG/DSC-200 with a heating rate of 10 °C min$^{-1}$ at the temperature range of 25 to 700 °C in a N$_2$ atmosphere. The surface area of carbon aerogel was determined from N$_2$ (77.4 K) adsorption–desorption isotherms using an ASAP-2046 surface area analyzer. The surface area was calculated using a Brunauer–Emmett–Teller (BET) method in the linear range of P/P$_0$=0.01–0.1. X-ray diffraction (XRD) patterns were recorded on a Bruker D8 diffractometer using Cu Kα radiation as the X-ray source. Infrared (IR) data were recorded on a Fourier transform IR spectrometer (VERTEX 70, Bruker Corp., Germany). Raman spectra were carried out on Raman spectrometer (LabRAM ARAMIS-Horiba Jobin Yvon) operating under 532 nm excitation. Compression, elasticity, and fatigue
resistance were performed on a compressive instrument (Instron 5565). The electrical current was measured on an electrochemical workstation (CHI 660E).

**Assembly and testing of sensor.** The C-HPMC/rGO-6-based sensor was assembled by placing C-HPMC/rGO-6 into two pieces of PET substrates adhered with Al sheets. The strain and loading pressure were conducted by using Instron 5565. The real-time current was recorded on CHI 660E (applying a voltage of 1 V).
S2. Figures for characterization, experimental results

**Figure S1.** Transmission electron microscopy (TEM) image of GO.
Figure S2. Scanning electron microscopy (SEM) images of A-HPMC.
Figure S3. Digital photographs of (a) A-HPMC and C-HPMC. (b) Stress-strain curves of A-HPMC.
Figure S4. (a) Digital photograph, (b) SEM image, and (c) stress-strain curves of C-R-HPMC/rGO-6 (fabricated by random freeze-casting).
Figure S5. The schematic diagram of directional freezing.
Figure S6. Thermogravimetric analysis (TGA) curves of the as-prepared aerogels.
Figure S7. (a) Nitrogen adsorption-desorption isotherms and (b) pore size distributions of C-HPMC/rGO-x.
Figure S8. FT-IR spectra of the as-prepared aerogels.
Figure S9. XRD patterns of A-GO and C-rGO.
Figure S10. Stress-strain curves of A-HPMC/GO-6.
Figure S11. Digital photographs of C-HPMC/rGO-6 with different solid contents after 10 cycles: (a) 5 wt%, (b) 7 wt%, and (c) 9 wt%, respectively.
Figure S12. Digital photographs of C-HPMC/rGO-x.
Figure S13. Stress-strain curves of (a) C-HPMC/rGO-1, (b) C-HPMC/rGO-2, (c) C-HPMC/rGO-10.
Figure S14. Mechanical properties of C-HPMC/rGO-x vs density.
S3. Tabulated results

**Table S1.** Stress retentions (30% strain) of C-HPMC/rGO-6 with different heating rates (300-400 °C).

<table>
<thead>
<tr>
<th>Heating rate (°C min⁻¹)</th>
<th>500ˢ (%)</th>
<th>1000ˢ (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.5</td>
<td>90.54</td>
<td>88.58</td>
</tr>
<tr>
<td>1.0</td>
<td>88.70</td>
<td>86.93</td>
</tr>
<tr>
<td>3.0</td>
<td>83.87</td>
<td>81.14</td>
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</tbody>
</table>
Table S2. Stress retentions of C-HPMC/rGO-6 with different treatment before carbonization at 30% strain.

<table>
<thead>
<tr>
<th>Process</th>
<th>500&lt;sup&gt;th&lt;/sup&gt; (%)</th>
<th>1000&lt;sup&gt;th&lt;/sup&gt; (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>With pre-Oxidation</td>
<td>94.73</td>
<td>94.10</td>
</tr>
<tr>
<td>Without pre-Oxidation</td>
<td>90.54</td>
<td>88.58</td>
</tr>
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</table>
**Table S3.** Stress retentions of C-HPMC/rGO-x with different mass rates of HPMC to GO at 50% strain.

<table>
<thead>
<tr>
<th>Cycles</th>
<th>100&lt;sup&gt;th&lt;/sup&gt;</th>
<th>500&lt;sup&gt;th&lt;/sup&gt;</th>
<th>1000&lt;sup&gt;th&lt;/sup&gt;</th>
</tr>
</thead>
<tbody>
<tr>
<td>1:1</td>
<td>87.58%</td>
<td>82.64%</td>
<td>-</td>
</tr>
<tr>
<td>2:1</td>
<td>87.20%</td>
<td>83.05%</td>
<td>-</td>
</tr>
<tr>
<td>10:1</td>
<td>Collapsed after 10 cycles</td>
<td></td>
<td></td>
</tr>
<tr>
<td>20:1</td>
<td>Collapsed after 1 cycles</td>
<td></td>
<td></td>
</tr>
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S4. References