Supporting Information

Freezing-tolerant, Widely Detectable and Ultra-sensitive Composite Organohydrogel for Multiple Sensing

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**Table S1** Preparation of different hydrogels.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Soaking solution</th>
<th>Soaking time (d)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pre-hydrogel (a–No Soaking)</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Organohydrogel–1 (b–1:1)</td>
<td>0.3 1:1</td>
<td>3</td>
</tr>
<tr>
<td>Organohydrogel–2 (c–2:1)</td>
<td>0.3 2:1</td>
<td>3</td>
</tr>
<tr>
<td>Hydrogel (d–1:0)</td>
<td>0.3 1:0</td>
<td>3</td>
</tr>
</tbody>
</table>
**Figure S1.** Schematic illustration of the sandwiched hydrogel strain sensor structure.

**Figure S2.** Schematic illustration of the sandwiched hydrogel pressure sensor structure.
Figure S3. (a) Digital image of four different samples by placing them under different environments: room temperature, -20 °C for one hour, -40 °C for one hour, -40 °C for three hours. (b) Photographs of hydrogels (a–No Soaking) twisted at room temperature and frozen after storage for 24 h at -20°C. (c) Photographs of hydrogels (b–1:1) are twisted at room temperature and after storage for 24 h at -20 °C. (d) Photographs of hydrogels (b–1:1) is twisted at room temperature and after storage for 3 h at -40°C.
Figure S4. DSC results of hydrogels.
Figure S5. Mechanical properties of the PDA–rGO/SA/PAM composite hydrogel.
Photos of hydrogel: (a) stretching after knotting and twisting, (b) bearing the pressure of a knife and a blunt-edged scissor, and (c) compressing.

Figure S6. Strain-stress cyclic curves of (a) a–No Soaking, (b) b–1:1, (c) c–2:1 and (d) d–1:0 hydrogels.
Figure S7. The real-time resistance variation (a) and sensitivity (b) at different strains (50%, 100%, 150%, 200%, 250%).
Figure S8. No irritation on human skin was detected after attaching the hydrogel for 4h indicating the hydrogel is safe to human skin.
**Figure S9.** The fitting curve and cubic function relationship between the relative resistance change ($\Delta R/R_0$) and bending angles.

\[
y = a\theta^3 + b\theta^2 + c\theta + d
\]

\[
a = 1.46843E-4 \quad b = -0.01969
\]

\[
c = 1.1406 \quad d = 1.42109E-14
\]

**Figure S10.** Response and release behavior of the strain sensor as the index finger bend.
**Figure S11.** Schematic for a situation of five sensors (thumb, index, middle, ring, little).

**Figure S12.** (a) Schematic for a situation of five sensors on the ball. (b) The real-time resistance changes corresponding to three, four, and five fingers pressing the ball.
Table S2. Comparison in the properties of hydrogel-based sensors based on different materials.

<table>
<thead>
<tr>
<th>Flexible sensor composition</th>
<th>Tensile strength (kPa)</th>
<th>Sensitivity (strain, GF)</th>
<th>Temperatur e sensing range (°C)</th>
<th>Anti-freezing properties</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>PDA–rGO/SA/PAM organohydrogel–1</td>
<td>143.2</td>
<td>0-250%, 2.09</td>
<td>-20 ~ 60</td>
<td>-20 °C, 24 h; -40 °C, 3 h</td>
<td>This work</td>
</tr>
<tr>
<td>PANI NFs/ PAA/Fe^{3+}</td>
<td>35.68</td>
<td>0-150%, 1.16</td>
<td>40 ~ 110</td>
<td>No</td>
<td>ACS Nano, 2020 1</td>
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<tr>
<td>PVA/Gly/CB/CNT</td>
<td>4800</td>
<td>0-700%, 2.01</td>
<td>30 ~ 80</td>
<td>-20 °C, 24 h</td>
<td>ACS Appl Mater Interfaces, 2020 2</td>
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<tr>
<td>PAM/carrageenan Gly-organohydrogels</td>
<td>36</td>
<td>Not given</td>
<td>25~ 102</td>
<td>-18 °C, 24 h</td>
<td>ACS Appl Mater Interfaces, 2020 3</td>
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<tr>
<td>PAAm/SA/CNT/CA Cl₂</td>
<td>271.68 ± 6.04</td>
<td>0-400%, 3.125</td>
<td>No</td>
<td>-20 °C</td>
<td>ACS Appl Mater Interfaces, 2020 4</td>
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<tr>
<td>Gelatin/PAAm-oxCNTs</td>
<td>710</td>
<td>0-250%, 1.50</td>
<td>No</td>
<td>No</td>
<td>Chemical Engineering Journal, 2020 5</td>
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<tr>
<td>PAA/CS/GO/Gly</td>
<td>226.2 ± 30.05</td>
<td>0-80%, 1.138</td>
<td>No</td>
<td>-20 °C</td>
<td>Journal of Materials Chemistry C, 2019 6</td>
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</tbody>
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References


