Supporting Information

Rational Design of Flexible Capacitive Sensors with Highly Linear Response over a Broad Pressure Sensing Range

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* Equal contribution to this work.
Fig. S1 The SEM image of spiky Ni particles.

Fig. S2 Fabrication of the dielectric layer with microstructure prepared from calathea zebrine leaf or abrasive paper.
**Fig. S3** Experimental and calculated compression stress (pressure)-strain (thickness change) curves of the 29.0 vol% spiky Ni/PDMS composite.

**Fig. S4** Dielectric constant (blue) and loss factor (red) of spiky Ni/PDMS composites as function of filler volume percentage. The black curve is a fitting curve of percolation theory.

**Fig. S5** (a) The actual change of capacitance as a function of the applied pressure. (b) The loss factor as a function of applied pressure.
Fig. S6 (a) The SEM images of spherical Ni particles. (b) The change of capacitance as a function of the applied pressure. (c) The percolation curve of the spherical Ni/PDMS dielectric layer.

Fig. S7 The loss factor as a function of applied pressure, including data from the composites with 21.4 vol.%, 23.1 vol.%, 26 vol.%, 27.8 vol.%, 29.0 vol.% spiky Ni and pure PDMS.

Fig. S8 The change of capacitance as a function of the applied pressure, and the linear fitting of the sensitivity curves of two different composites.
Fig. S9 Digital photos of the traditional flexible film covering at the bottom of the flask.

Fig. S10 Digital photos of the sensor covering at curved surface.
The change of capacitance as a function of the applied pressure at lower than 4 kPa. Since the air pressure changes 1.2 kPa when the altitude increases by 100 meters, the equivalent capacitance change is approximately 0.0672. That is, the linear coefficient between altitude and capacitance change is $100/0.0672=1488$.

**Table S1.** The characterization of the dielectric layer composites (calculated).

<table>
<thead>
<tr>
<th>Sample</th>
<th>Fitted Curve</th>
</tr>
</thead>
<tbody>
<tr>
<td>21.4 vol% spiky Ni</td>
<td>$y=0.0018x+0.0374, R^2=0.9633$</td>
</tr>
<tr>
<td>23.1 vol% spiky Ni</td>
<td>$y=0.002x+0.0401, R^2=0.9680$</td>
</tr>
<tr>
<td>26.0 vol% spiky Ni</td>
<td>$y=0.0028x+0.0452, R^2=0.9804$</td>
</tr>
<tr>
<td>27.8 vol% spiky Ni</td>
<td>$y=0.004x+0.0456, R^2=0.9918$</td>
</tr>
<tr>
<td>29.0 vol% spiky Ni</td>
<td>$y=0.0058x+0.0337, R^2=0.9951$</td>
</tr>
<tr>
<td>31.0 vol% spiky Ni</td>
<td>$y=0.0085x+0.0264, R^2=0.9844$</td>
</tr>
</tbody>
</table>

**Table S2.** The characterization of the dielectric layer composites (experimental).

<table>
<thead>
<tr>
<th>Sample</th>
<th>Fitted Curve</th>
</tr>
</thead>
<tbody>
<tr>
<td>pure PDMS</td>
<td>$y=0.0002x+0.1741, R^2=0.7622$</td>
</tr>
<tr>
<td>21.4 vol% spiky Ni</td>
<td>$y=0.0007x+0.3529, R^2=0.8706$</td>
</tr>
<tr>
<td>23.1 vol% spiky Ni</td>
<td>$y=0.0009x+0.4572, R^2=0.8600$</td>
</tr>
<tr>
<td>26.0 vol% spiky Ni</td>
<td>$y=0.001x+0.5094, R^2=0.8499$</td>
</tr>
<tr>
<td>27.8 vol% spiky Ni</td>
<td>$y=0.0022x+0.3753, R^2=0.9857$</td>
</tr>
<tr>
<td>29.0 vol% spiky Ni</td>
<td>$y=0.0046x+0.3208, R^2=0.9985$</td>
</tr>
<tr>
<td>31.0 vol% spiky Ni</td>
<td>$y=0.0053x+0.7706, R^2=0.9665$</td>
</tr>
</tbody>
</table>
**Table S3.** Typical sensitivities, working ranges and linearity of previously reported linear flexible capacitive pressure sensors.

<table>
<thead>
<tr>
<th>Materials</th>
<th>Sensitivity (kPa(^{-1}))</th>
<th>Detection range (kPa)</th>
<th>S×LMR</th>
<th>Linearity</th>
<th>Journal</th>
<th>Ref</th>
</tr>
</thead>
<tbody>
<tr>
<td>our work-flat surface</td>
<td>0.0046</td>
<td>0-1700</td>
<td>7.8</td>
<td>0.999</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>our work-with leaf surface structure</td>
<td>0.0216</td>
<td>0-500</td>
<td>10.8</td>
<td>0.9978</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>our work-with abrasive paper surface structure</td>
<td>1.149</td>
<td>0-20</td>
<td>23.0</td>
<td>0.9989</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>silver nanowires/PDMS</td>
<td>0.00016</td>
<td>0-500</td>
<td>0.08</td>
<td>-</td>
<td>Nanoscale [1]</td>
<td></td>
</tr>
<tr>
<td>carbon black/silicone rubber</td>
<td>0.0002536</td>
<td>0-700</td>
<td>1.65</td>
<td>0.9981</td>
<td>Meas. Sci. Technol. [2]</td>
<td></td>
</tr>
<tr>
<td>PDMS/CIP with a hair-like micro cilia array structure</td>
<td>0.28</td>
<td>0-10</td>
<td>2.8</td>
<td>0.981</td>
<td>J. Mater. Chem. A [3]</td>
<td></td>
</tr>
<tr>
<td>microstructural single-walled carbon nanotubes/PDMS</td>
<td>0.7</td>
<td>0-25</td>
<td>17.5</td>
<td>-</td>
<td>Adv. Mater. [4]</td>
<td></td>
</tr>
<tr>
<td>porous carbon black/PDMS</td>
<td>1.1</td>
<td>0-10</td>
<td>11.0</td>
<td>-</td>
<td>Nanotechnolog [5]</td>
<td></td>
</tr>
</tbody>
</table>
Table S4. Typical sensitivities and working ranges of recently capacitive pressure sensors

<table>
<thead>
<tr>
<th>Materials</th>
<th>Sensitivity (kPa⁻¹)</th>
<th>Detection range (kPa)</th>
<th>Journal</th>
<th>Ref</th>
</tr>
</thead>
<tbody>
<tr>
<td>PDMS dielectric layer with hollow micro-pillars</td>
<td>0.0014 (10-20 kPa)</td>
<td>10-120</td>
<td>Nano Energy</td>
<td>[6]</td>
</tr>
<tr>
<td></td>
<td>0.0005 (20-120 kPa)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>MXene/PVDF-TrFE composite nanofibrous scaffolds as a dielectric layer</td>
<td>0.51 (0-1 kPa)</td>
<td>0-400</td>
<td>ACS Appl. Mater.</td>
<td>[7]</td>
</tr>
<tr>
<td>between PEDOT:PSS electrodes</td>
<td>0.011 (10-150 kPa)</td>
<td></td>
<td>Inter.</td>
<td></td>
</tr>
<tr>
<td></td>
<td>0.006 (150-400 kPa)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>PDMS pyramids</td>
<td>0.55</td>
<td>0-7</td>
<td>Nat. Mater.</td>
<td>[8]</td>
</tr>
<tr>
<td>polyurethane/multiwalled carbon nanotubes</td>
<td>0.753 (&lt; 2 kPa)</td>
<td>0.1-50</td>
<td>ACS Appl. Mater.</td>
<td>[9]</td>
</tr>
<tr>
<td></td>
<td>0.0549 (2-50 kPa)</td>
<td></td>
<td>Inter.</td>
<td></td>
</tr>
<tr>
<td>PDMS film with uniformly distributed micro-pores as a dielectric layer</td>
<td>1.18</td>
<td>0-0.02</td>
<td>Sensor. Actuat.</td>
<td>[10]</td>
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<tr>
<td></td>
<td></td>
<td>A-Phys.</td>
<td></td>
<td></td>
</tr>
<tr>
<td>AgNWs/PDMS as the electrode, while PVDF as the dielectric layer</td>
<td>2.94 (0-1.5 kPa)</td>
<td>0-7</td>
<td>ACS Appl. Mater.</td>
<td>[11]</td>
</tr>
<tr>
<td></td>
<td>0.75 (1.5-7 kPa)</td>
<td></td>
<td>Inter.</td>
<td></td>
</tr>
<tr>
<td>AgNWs/PDMS as the electrode, while PVP or PMMA as the dielectric layer</td>
<td>3.8 (0.045-0.5 kPa)</td>
<td>0.045-5</td>
<td>Nanoscale</td>
<td>[12]</td>
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<tr>
<td></td>
<td>0.8 (0.5-2 kPa)</td>
<td></td>
<td></td>
<td></td>
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<tr>
<td></td>
<td>0.35 (2-5 kPa)</td>
<td></td>
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<tr>
<td>porous film loading with ionic liquid on the fabric skeleton, then coated</td>
<td>4.46 (&lt; 0.5 kPa);</td>
<td>0-120</td>
<td>Mater. Today</td>
<td>[13]</td>
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<tr>
<td>with AgNWs</td>
<td>0.5 (0.5-10 kPa)</td>
<td></td>
<td>Phys.</td>
<td></td>
</tr>
<tr>
<td></td>
<td>0.0143 (10-120 kPa)</td>
<td></td>
<td></td>
<td></td>
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<tr>
<td>PVDF-TrFE interlocked microstructure</td>
<td>6.583 (0-0.1 kPa)</td>
<td>0-0.9</td>
<td>Small</td>
<td>[14]</td>
</tr>
<tr>
<td></td>
<td>0.125 (0.1-0.9 kPa)</td>
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<td></td>
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<td>PDMS pyramids and Polyisoindigobithiophene-siloxane transistor</td>
<td>8.4 (0-8 kPa)</td>
<td>0-60</td>
<td>Nat. Commun.</td>
<td>[15]</td>
</tr>
<tr>
<td></td>
<td>0.38 (8-60 kPa)</td>
<td></td>
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</tbody>
</table>

**Video S1.** The response of the sensor during it descends and ascends in water at certain speed.
References