Supporting Information

**Large-Area Superelastic Graphene Aerogels Based on Room-Temperature Reduction Self-Assembly Strategy for Sensing and Particulate Matter (PM\textsubscript{2.5} and PM\textsubscript{10}) Capture**

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![Figure S1](image1.png)

**Figure S1.** Digital photos of differently shaped RGHs depending on different kinds of reactors: a) tubular; b) membranous; and c) cylindrical.

![Figure S2](image2.png)

**Figure S2.** The volume shrinkage of GA as a function of concentration of GO.
Figure S3. Digital images of GAs with different experimental parameters: GO concentration, reduction time, pre-freeze temperature and pre-freeze time.

Figure S4. The volume shrinkage of GA as a function of a) reduction time (self-assembly time), b) pre-freeze time and c) pre-freeze temperature, respectively.

Table S1. The effect of pH value of GO mixture with different reducing agents on the formation of RGH.

<table>
<thead>
<tr>
<th>Reducing agent</th>
<th>RGH forming conditions (pH value)</th>
<th>Whether RGH is obtained</th>
</tr>
</thead>
<tbody>
<tr>
<td>HI</td>
<td>—</td>
<td>No</td>
</tr>
<tr>
<td>LAA</td>
<td>7-9</td>
<td>Yes</td>
</tr>
<tr>
<td>Hydrazine hydrate</td>
<td>8-11</td>
<td>Yes</td>
</tr>
<tr>
<td>NaBH₄</td>
<td>10-11</td>
<td>Yes</td>
</tr>
<tr>
<td>EDA</td>
<td>—</td>
<td>No</td>
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</tbody>
</table>
Figure S5. a) Photographs illustration of the fabrication process of GA. b) The cross-sectional SEM image of GA prepared from 5 mg ml\(^{-1}\) GO dispersions. c) Photographs illustration of the GA under one compression cycle. The GA were prepared without adding ammonia into the GO dispersions.

Figure S6. Photographs of room-temperature self-assembly of GO dispersions with different PH value: a-c) PH=1, 3 and 5, respectively. The self-assembly of GO was conducted by using the hydrazine hydrate as the reducing agent.
Figure S7. The BJH pore size distribution of GAs with different density.

Figure S8. XRD patterns of GO and the GA.

Figure S9. Full range survey scan of GO and the GA.
Figure S10. Digital images show the volume shrinkage of RGH and compressibility of GA prepared with different reducing agents: a-b) RGH and GA prepared with LAA as the reducing agent; c-d) RGH and GA prepared with NaBH₄ as the reducing agent.

Figure S11. FTIR spectrum of GO and the GA.
Figure S12. SEM images of the microstructures of GAs after capturing PM particles and washing with water: a) after one capture-washing cycle; b) after four capture-washing cycles.

Legends for Supplementary Movies

Movie S1: Movie S1 illustrates the brightness of the LED changed with the compressing and releasing processes.

Movie S2: Movie S2 illustrates the PM removal process of the prepared GA.