Controlling the cooperative self-assembly of graphene oxide quantum dots in aqueous solutions

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Preparation and characterization of GOQDs

Figure S1. The synthetic scheme for preparation of the GOQDs
Figure S2. a) DSC analysis of the GOQDs in N₂ atmosphere, b) TGA (solid line) and DTG (dots line) curves in N₂ atmosphere.
**Figure S3.** Raman spectra of Synthesized GOQD

**Particle sizes for GOQD solutions**

**Table S1.** Average sizes of the GOQD and GOQD assemblies at different concentrations in deionized H$_2$O as determined by DLS at 25 °C.

<table>
<thead>
<tr>
<th>GOQD [mg/ml]</th>
<th>Average size in diameter [nm]</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.01</td>
<td>0.7 +/- 0.2</td>
</tr>
<tr>
<td>0.02</td>
<td>0.9 +/- 0.1</td>
</tr>
<tr>
<td>0.05</td>
<td>1.2 +/- 0.7</td>
</tr>
<tr>
<td>0.067</td>
<td>66.7 +/- 4.1</td>
</tr>
<tr>
<td>0.1</td>
<td>66.7 +/- 9.4</td>
</tr>
<tr>
<td>1.0</td>
<td>78.3 +/- 19</td>
</tr>
</tbody>
</table>

**Table S2.** Average sizes of the GOQDs and GOQD assemblies at a concentration of 0.05 mg/ml after addition of HNO$_3$ (pH 2.15), only deionized H$_2$O (pH 6.93) and after addition of NaOH (pH 11.9) as determined by DLS at 25 °C.

<table>
<thead>
<tr>
<th>pH</th>
<th>Average size in diameter [nm]</th>
</tr>
</thead>
<tbody>
<tr>
<td>2.1</td>
<td>73.1 +/- 9.42</td>
</tr>
</tbody>
</table>
Zeta potentials ($\zeta$) of GOQD solutions

Figure S4. Zeta potential ($\zeta$) measurements of the GOQD solution in the 0.05 mg/ml (a) and 0.1 mg/ml (b) concentrations at deionized water.

TEM images
Figure S5. HR-TEM images of the GOQDs at concentration a) under CAC (0.05 mg/ml) and b) above CAC (1 mg/ml).

UV-Vis transmittance

Figure S6. Typical representation of the UV-Vis transmittance of 1 mg/ml GOQDs solution.
AFM images and size analysis in the dried state

**Figure S7.** Approximate size and thickness of the single layer and self-assembled GOQDs through AFM images as a function of pH determined by DLS at 25 °C. With HNO₃ addition (pH=2.1), no addition (pH=6.9) and with NaOH (pH=11.9) at a concentration of 0.05 mg/ml in deionized H₂O.

**Fluorescence spectroscopy**
Figure S8. Fluorescence of 0.05 mg/ml (blue) and 1 mg/ml (red) solutions of GOQDs in deionized H$_2$O at different excitations and emissions.