Supporting Information

Optical and Electronic Properties of Graphene Quantum Dots with Oxygen-Containing Groups: A Density Functional Theory Study

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Fig. S1 Structures of C24, C32, and C42 and their functionalized GQDs.
Table S1: Absorption energies, wavelengths, and oscillator strengths for the first 20 singlet states. The values were obtained using B3LYP/6-31G* for the optimized ground state geometry. Water was taken into account as solvent by using the polarizable continuum model.

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**Table S6:** Absorption energies, wavelengths, and oscillator strengths for the first 20 singlet states of GQD-OH8-EF.

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**Table S7:** Absorption energies, wavelengths, and oscillator strengths for the first 20 singlet states of GQD-COOH2-SF.

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Table S8: Absorption energies, wavelengths, and oscillator strengths for the first 20 singlet states of GQD-CHO2-SF.

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**Table S9**: Absorption energies, wavelengths, and oscillator strengths for the first 20 singlet states of GQD-COC2-SF.

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Table S10: Absorption energies, wavelengths, and oscillator strengths for the first 20 singlet states of GQD-OH2-SF.

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Table S11: Absorption energies, wavelengths, and oscillator strengths for the first 20 singlet states of GQD-OCH$_3$2-SF.

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Fig. S2 Calculated absorption spectra of C24-C42 and their functionalized GQDs.
Fig. S3 Isosurfaces of HOMO and LUMO in GQDs with different oxygen-containing groups in ground state. The positive and negative orbital lobes are displayed in red and green, respectively. (a) C132-CHO8-EF, (b) C132-OCH38-EF, (c) C132-OH8-EF, (d) C132-CHO2-SF, (e) C132-OCH32-SF, (f) C132-OH2-SF.
**Fig. S4** Structures of edge-functionalized GQDs with carboxyl and epoxy groups.

**Fig. S5** Calculated absorption spectra of edge-functionalized GQDs with carboxyl and epoxy groups in Fig. S4.
Fig. S6 Schematics of surface functionalization position on C132 GQDs: (a) C132-R2-SF-1, (b) C132-R2-SF-2, (c) C132-R2-SF-3, and (d) C132-R2-SF-4. R indicates the functional group.
Fig. S7 Calculated absorption spectra of the surface-functionalized GQDs with five oxygen groups in Fig. S6.
Table S12: HOMO energy, LUMO energy, HOMO-LUMO gap, and absorption maximum wavelengths of the surface-functionalized GQDs with different positions.

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Fig. S8 Isosurfaces of HOMO and LUMO in GQDs with different surface positions in ground state. (a) C132-COOH2-SF-1, (b) C132-COOH2-SF-2, (c) C132-COOH2-SF-3, (d) C132-COOH2-SF-4.
Table S13: Excitation energies, wavelengths, oscillator strengths, transition coefficients, and associated eigenvalues of the dominated excitation in GQDs.

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<th>Transition coefficients</th>
<th>Associated eigenvalues( $\lambda_i$ )</th>
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<td>C132-OH2-SF</td>
<td>S8</td>
<td>1.68</td>
<td>736.51</td>
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<td>H-5 -&gt; L</td>
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**Fig. S9** Natural transition orbital pairs for the dominant excited state of GQDs. (a) C132-CHO8-EF, (b) C132-OCH38-EF, (c) C132-OH8-EF, (d) C132-CHO2-SF, (e) C132-OCH32-SF, (f) C132-OH2-SF. For this state the “hole” is on the left, the “particle” is on the right; The values represent the associated eigenvalue ($\lambda_i$) of respective NTOs.
Fig. S10 Representation of the electron difference density between the dominated excited state minus ground state for GQDs. (a) C132-CHO8-EF, (b) C132-OCH₃8-EF, (c) C132-OH8-EF, (d) C132-CHO2-SF, (e) C132-OCH₂2-SF, (f) C132-OH2-SF. At the bottom, the lateral views are shown. The blue area plots the surface where the value of the difference density is -0.0002 and the yellow area plots the surface where the value of the difference density is +0.0002.