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Graphene quantum dots with controllable surface oxidation, tunable fluorescence and up-conversion emission

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Quantum yields (QY) measurement.

9,10-Bis (phenylethynyl) anthracene in cyclohexane (QY=1) was chosen as standard. The quantum yields of s-GQDs (in water) were calculated according to:

$$\phi_x = \phi_{st}(I_x / I_{st})(\eta_x^2 / \eta_{st}^2)(A_{st} / A_x)$$

Where φ is the quantum yield, I is the integrated emission intensity, η is the refractive index of the solvent, and A is the optical density. The subscript *"st"* refers the to standard with known quantum yield and *"x"* refers to the sample. To minimize re-absorption effects, absorption in the 10 mm fluorescence cuvette was kept below 0.10 at the excitation wavelength (425 nm).

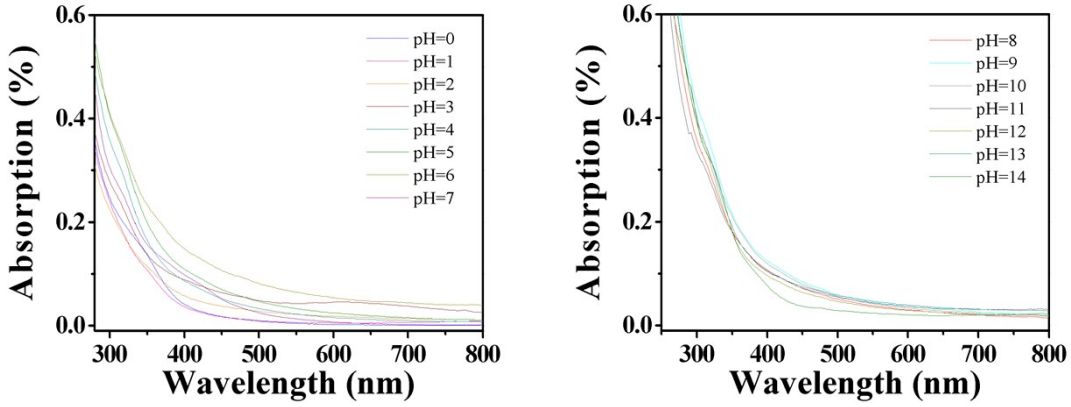
Table S1: Quantum yield of s-GQDs using 9,10-bis (phenylethynyl) anthracene as a reference.

Sample	Integrated emission intensity *(I)*	Abs. at 425 nm *(A)*	Refractive index of solvent *(η)*	Quantum Yields *(φ)*
9,10-Bis (phenylethynyl) anthracene	20198.1	0.069	1.4264	1 (known)
Batch 1	289.3	0.021	1.33	0.041
Batch 2	1327.8	0.040	1.33	0.099
Batch 3	942.1	0.023	1.33	0.122

Note: The "UCPL" in GQDs may be due to the excitation of second-order diffraction light (wavelength $\lambda/2$), which coexists in the selected first-order light (wavelength λ) from the monochromators in the fluorescence spectrophotometer (See "Graphene Science Handbook,

CRC, 163-178", Section 10.4 Is There Real Up-Conversion PL from GQDs? and relative reference[1]) Thus, the up-conversion emission of GQDs and true "two-photon excited emission" in bioimaging should be carefully reinvestigated in detail.

Fig. S1 UV-vis absorption (ABS) spectra of the GQDs (Batch 3) in different pH solutions.



1. Z. Gan, X. Wu, G. Zhou, J. Shen and P. K. Chu, *Advanced Optical Materials*, 2013, **1**, 554-558.