## **Supplementary Information**

## Green Preparation of High Yield Fluorescent Graphene Quantum Dots from Coal-Tar-Pitch by Mild Oxidation

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Sample	Elements (%)				
	С	0	Ν	S	
CTP	93.54	3.58	2.62	0.26	
GQDs-1	69.30	29.84	0.77	0.09	
GQDs-2	58.85	40.10	0.98	0.07	

 Table S1. Summary of surface element content of CTP and GQDs by XPS analysis.

Table S2. Summary of C chemical bonds form in the surface of CTP and GQDs.

Chemical bond type	Chemical bond proportion (%)			
	CTP	GQDs-1	GQDs-2	
Csp <sup>2</sup>	51.30	37.30	30.98	
Csp <sup>3</sup>	39.49	23.59	20.35	
C-0	9.21	12.04	8.00	
C=O	-	5.86	18.28	
СООН	-	21.21	22.37	



Fig. S1. Macroscale image and simplified illustrative molecular structure of CTP.



**Fig. S2.** Characterization of CTP. (a) XPS survey of CTP. (b) High resolution C 1s XPS spectrum of CTP displayed presence of Csp<sup>2</sup>, Csp<sup>3</sup> and C–O modes at 284.6 eV, 285.1 eV and 285.4 eV, respectively. (c) ssFTIR spectrum of CTP showing H–Csp<sup>2</sup>, C–O, C=C, H–Csp<sup>3</sup> and O–H vibration modes at 750/3040 cm<sup>-1</sup>, 1032 cm<sup>-1</sup>, 1593 cm<sup>-1</sup>, 2917 cm<sup>-1</sup> and 3426 cm<sup>-1</sup> as labeled, respectively. (d) Raman spectrum of CTP under excitation of 532 nm.



**Fig. S3.** TEM image of CTP after ultrasonication in hydrogen peroxide for 2 h. Inset is the optical image.



Fig. S4. Preparation procedure of GQDs-1.



Fig. S5. The height distribution of the GQDs-1.



**Fig. S6.** Characterization of GQDs-2. (a) TEM image of GQDs-2 displaying a regular size and shape distribution. Scale bar, 20 nm. (b) The size distribution histogram of GQDs-2. (c) HRTEM image of representative GQDs-2 from a. (d) AFM image of GQDs-2.



Fig. S7. The height distribution of the GQDs-2.



**Fig. S8.** Characterization of GQDs-2. (a) XPS survey of GQDs-2. (b) High-resolution C 1s XPS spectrum of GQDs-2. (c) The high-resolution O 1s XPS spectrum of GQDs-2. (d) ssFTIR spectrum of GQDs-2 showing different vibration modes as labeled.



**Fig. S9.** Optical properties of the GQDs-2. (a) UV-Visible spectrum of the GQDs-2 dispersed in water. Inset of panel a: the left is a photograph of the corresponding GQDs-2 aqueous solution under UV light with 365 nm excitation; the right is a photograph of the corresponding GQDs-2 aqueous solution taken under visible light. (b) PL spectra of the GQDs-2 solution under different excitation wavelengths.



**Fig. S10.** PLE spectrum of GQDs-2 with the detection wavelength of 430 nm and PL spectrum excited at 328 nm.