Electronic Supplementary Material (ESI) for Analytical Methods.

Supporting Information

Post-synthetic regulating the fluorescence of CDs: giving insight into the fluorescence mechanism

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Supplementary Note 1. Synthesis of pre-prepared nitrogen-free CDs.

Pre-prepared amino free CDs were synthesized according to the methods mentioned in the literature[1] with slight modifications. Typically, phloroglucinol (0.5g) was mixed with 10mL of ethanol. After10min of sonication, the clear solution was transferred into a 25mL Teflon-lined autoclave, followed by adding 2mL of HCl (37%) as the catalyst. The mixture was heated at 200℃ for 9h.

Supplementary Note 2. Post-synthetic regulating the fluorescence of nitrogen-free CDs.

Take 2ml of triphenylphosphine DMF solution with a mass fraction of 6wt% into the cuvette, add 40μL of pre-prepared CDs solution with pipette gun, mix evenly, seal the cuvettes with plug and sealing glue. Finally, the cuvette was placed into ultraviolet lamp (365nm) at 10cm and irradiated at different radiation times (0-4h) for spectral detection.

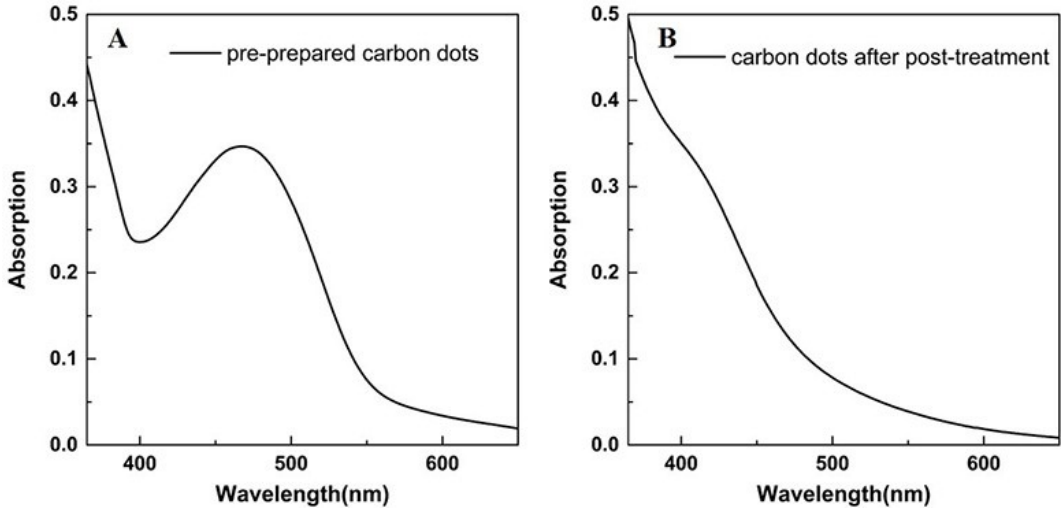

Figure S1. Absorption spectra of the pre-prepared CDs (A) and the CDs after post-treatment (B).

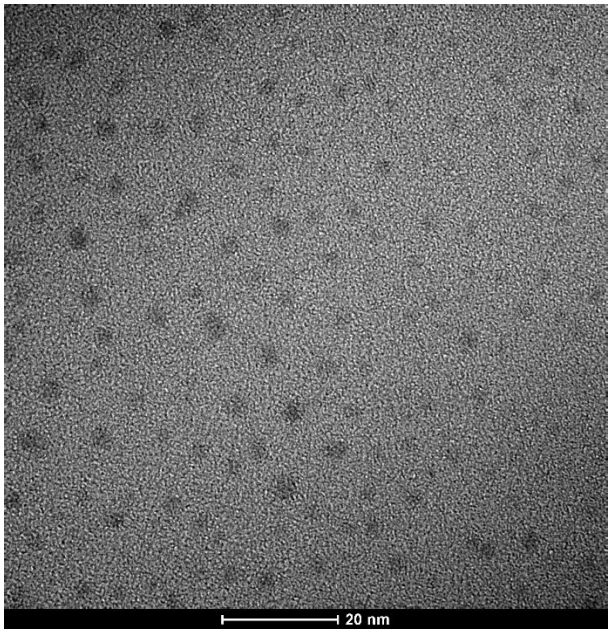

Figure S2. Transmission electron microscopy (TEM) image of pre-prepared CDs.

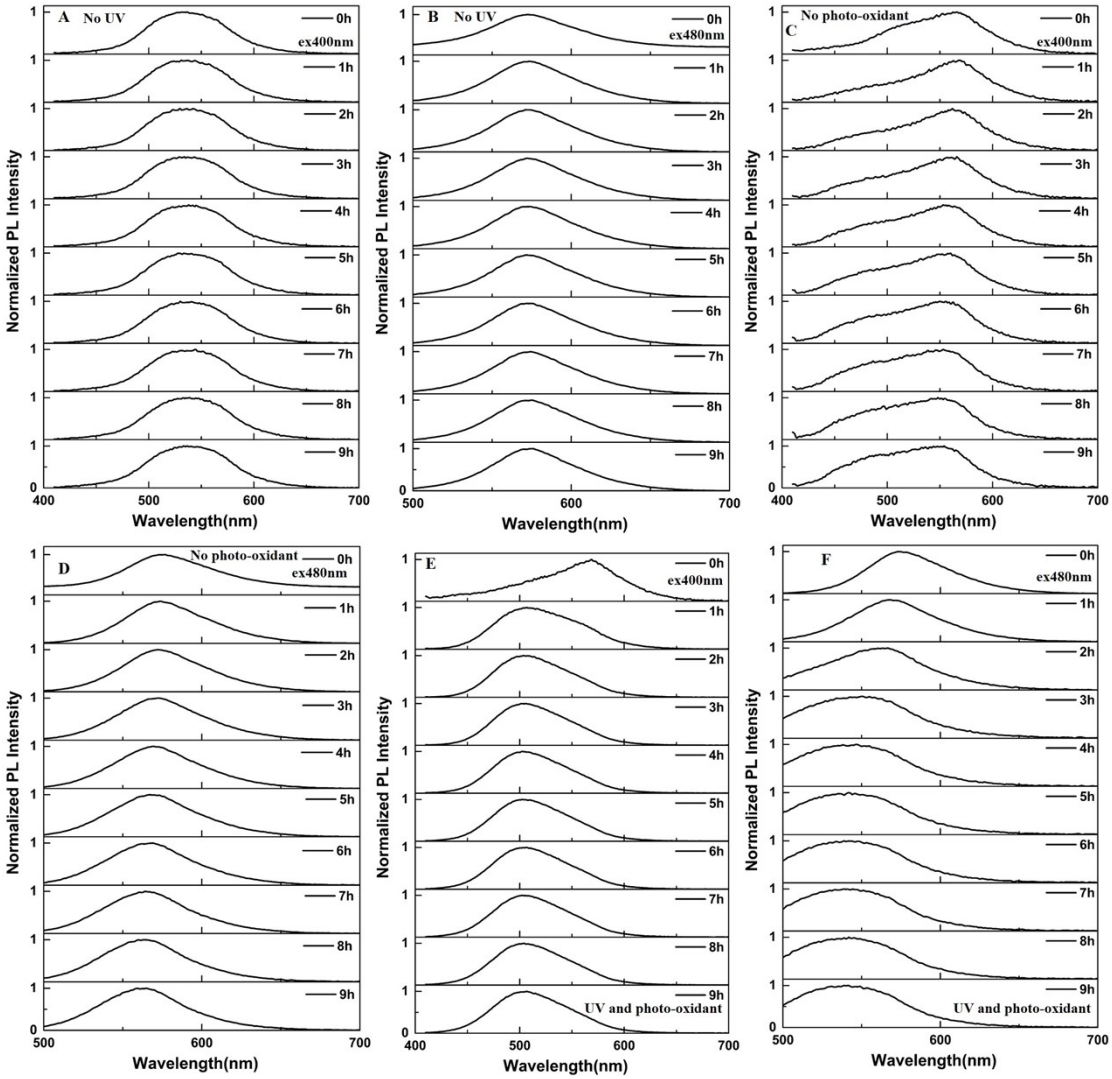

Figure S3. The normalized PL intensity of CDs when fluorescence regulation without photo-oxidant or UV. (A and B) The normalized PL intensity of CDs at excitation wavelengths of 400nm (A) and 480nm (B) in the presence of photo-oxidant but no UV irradiation (0-9h). (C and D) The normalized PL intensity of CDs at excitation wavelengths of 400nm (C) and 480nm (D) in the presence of UV irradiation but no photo-oxidant (0-9h). (E and F) The normalized PL intensity of CDs at excitation wavelengths of 400nm (E) and 480nm (F) in the presence of UV irradiation and no photo-oxidant (0-9h).

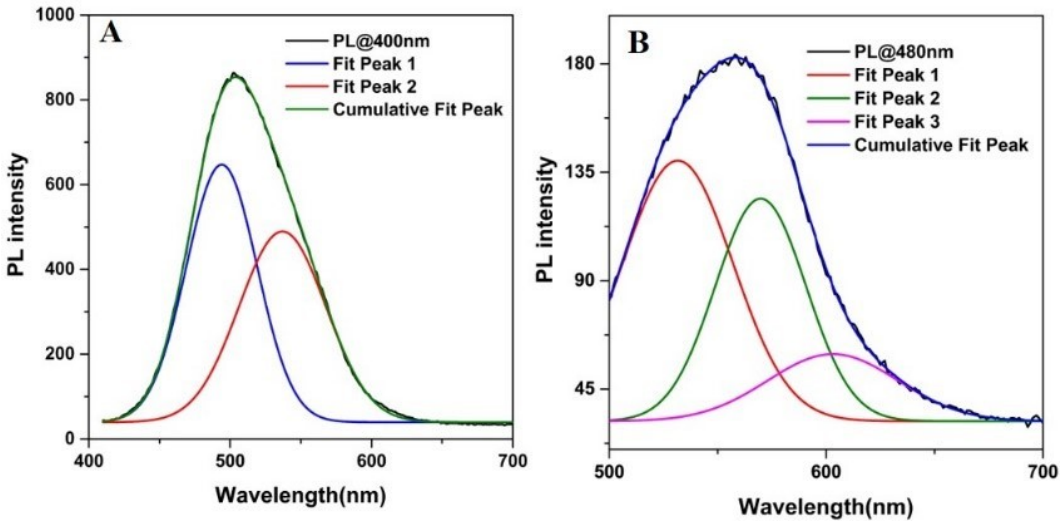

Figure S4. Sub-peak fitting strategy for CD emission. (A) The sub-peaks of PL were obtained by CDs at the excitation wavelength of 400nm: peak 1 (494nm) and peak 2 (537nm) under default conditions. (B)The sub-peaks of PL were obtained by CDs at the excitation wavelength of 480nm: peak 1 (537nm), peak 2 (560nm) and peak 3 (600nm) under default conditions.

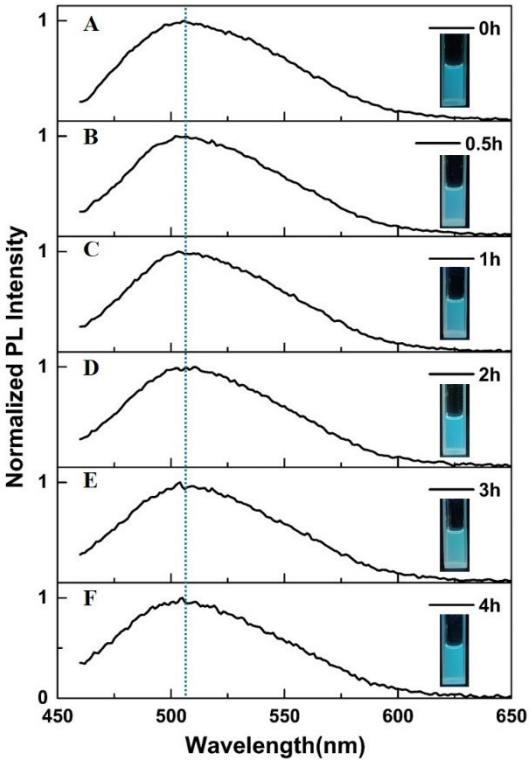

Figure S5. Optical performance of nitrogen-free CDs. (A to F) PL spectra of nitrogen-free CDs after post-treatment by 6wt% triphenylphosphine and UV irradiation for different times of 0-4h. The excitation wavelength is set as 450nm.

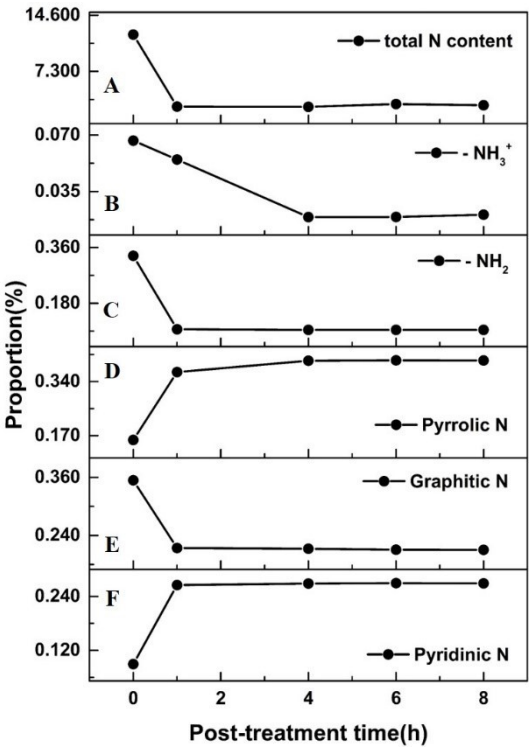

Figure S6. Dynamic content of components in high-resolution N1s XPS. (A) The total content of nitrogen atom under UV radiation 0-8h; (B to F) The proportion of each component of N1s under UV radiation 0-8h.

Table S1. The original data of the relative intensity ratios of various functional groups at different post-treatment times of 0, 1, 2, 3, 4, 6, and 8h (the -SCN bond is selected as the reference peak).

	C=O at 1750-1680cm^{-1}	C=O at 1650-1600cm^{-1}	-OH at 3450cm^{-1}	P=O at 1140-1250cm^{-1}	C-N at 1400cm^{-1}	C-N at 1223cm^{-1}	-NH_2 at 1655-1590cm^{-1}	-COOH at 1450-1410cm^{-1}
0h	0.004	0.115	0.275	0.039	1.424	0.944	0.532	0.254
1h	0.269	0.301	0.207	1.520	0.598	0.412	0.374	1.287
2h	0.345	0.348	0.314	1.593	0.543	0.311	0.341	1.360
3h	0.423	0.543	0.665	1.487	0.513	0.074	0.400	1.322
4h	0.370	0.506	0.357	1.383	0.559	0.182	0.310	1.278
6h	0.23	0.405	0.206	1.448	0.473	0.182	0.273	1.261
8h	0.217	0.565	0.361	1.690	0.523	0.273	0.325	1.360

Table S2. Normalize the relative intensity ratio between 0 and 8 h of each functional group from 0 to 1. To clearly observe the trend of the relative intensity ratio of each functional group, the following data can be plotted as Figure 3 B to I.

	C=O at 1750-1680cm^{-1}	C=O at 1650-1600cm^{-1}	-OH at 3450cm^{-1}	P=O at 1140-1250cm^{-1}	C-N at 1400cm^{-1}	C-N at 1223cm^{-1}	-NH_2 at 1655-1590cm^{-1}	-COOH at 1450-1410cm^{-1}
0h	0.000	0.000	0.150	0.000	1.000	1.000	1.000	0.000
1h	0.632	0.413	0.000	0.896	0.131	0.292	0.389	0.933
2h	0.814	0.518	0.235	0.941	0.073	0.212	0.263	1.000
3h	1.000	0.950	1.000	0.876	0.041	0.000	0.490	0.965
4h	0.872	0.869	0.329	0.813	0.089	0.111	0.142	0.926
6h	0.741	0.745	0.000	0.853	0.000	0.071	0.000	0.910
8h	0.707	1.000	0.337	1.000	0.052	0.163	0.198	0.999

Table S3. The proportion of each component of O1s under UV radiation 0h, 1h, 4h, 6h, 8h

	533.2eV -OH (%)	532.1eV C-O (%)	531.3eV C=O (%)
0h	0.227	0.388	0.383
1h	0.168	0.306	0.524
4h	0.245	0.157	0.597
6h	0.170	0.231	0.598
8h	0.177	0.232	0.590

Table S4. The proportion of each component of C1s under UV radiation 0h, 1h, 4h, 6h, 8h

	291.5eV C-C=O (%)	288.2eV C=O (%)	286.1eV C-O (%)	284.9eV C-N (%)	284.6eV C-C/C=C (%)
0h	0.000	0.040	0.335	0.428	0.195
1h	0.061	0.007	0.051	0.352	0.527
4h	0.055	0.014	0.032	0.355	0.542
6h	0.053	0.015	0.034	0.352	0.544
8h	0.054	0.014	0.032	0.350	0.546

Table S5. The proportion of each component of N1s under UV radiation 0h, 1h, 4h, 6h, 8h

	402.0eV $-NH_3^+$ (%)	401.1eV $-NH_2$(%)	400.5eV graphitic N (%)	399.6eV pyrrolic N (%)	398.9eV pyridinic N (%)
0h	0.066	0.333	0.353	0.156	0.089
1h	0.054	0.095	0.214	0.369	0.265
4h	0.019	0.093	0.212	0.405	0.268
6h	0.019	0.093	0.210	0.406	0.269
8h	0.020	0.093	0.210	0.406	0.269

References

1. H. Zhao, G. Liu and G. Han, *Nanoscale Advances*, 2019, **1**, 4888-4894.