Supplementary information

Rational Synthesis of Highly Efficient Ultra-narrow Red-emitting Carbon Quantum Dots For NIR-II Two-photon Bioimaging

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Materials

Reagent grade 1,1-Diphenyl-2-picrydrazyl (DPPH, 98.5%) was purchased from Macklin Biochemical Co., Ltd. (Shanghai, China). Reagent grade cumyl hydroperoxide (CHP), benzoyl peroxide (BPO), ammonium persulfate (APS), hydrogen peroxide (H_2O_2, 30% aqueous solution) and ferrous chloride tetrahydrate were purchased from Energy Chemical-Saan Chemical Technology Co., Ltd. (Shanghai, China). Reagent grade hydrobromic acid (HBr), sulfuric acid (H_2SO_4), phosphoric acid (H_3PO_4), nitric acid (HNO_3) and acetic acid (AcOH) were purchased from Aladdin Chemicals Co. Ltd (Shanghai, China).

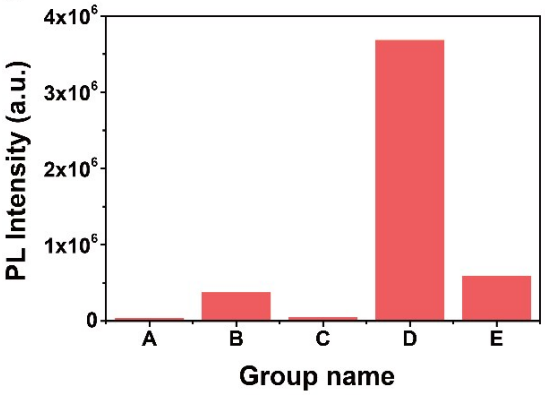
Methods

Control groups with different reacting temperature/ reaction time (Figure S2): T-CQDs were synthesized under identical condition described in the manuscript, except the reacting temperature or reaction time were changed.

Control groups with different additive acid/ oxidative radical reagents (Figure S3): T-CQDs were synthesized under identical condition described in the manuscript, except the additive acid or oxidative radical reagents were changed. (Fenton reagent used as oxidative radical reagent were prepared by mixing hydrogen peroxide and ferrous chloride at the molar ratio of 2:1)

Repeatability test groups: T-CQDs were synthesized under identical condition described in the manuscript for 5 batches, the first three batches involved 10 mL reacting solution in a 20 mL autoclave, the latter two batches involved 25 mL in a 50 mL autoclave and 100 mL in a 200 mL autoclave, respectively.

a



b

Group	HCl	TBHP	DPPH
A	✗	✗	✗
B	✓	✗	✗
C	✗	✓	✗
D	✓	✓	✗
E	✓	✓	✓

Figure S1 T-CQDs synthetic control groups. Group A: no acid or oxidative radical reagent; Group B: additive acid and no radical reagent; Group C: no acid, additive radical reagent; Group D: additive acid and radical reagent; Group E: additive acid and radical reagent, radical quencher DPPH was added. (a). The overall PL intensity measured in the five control groups. (b). The experimental condition applied.

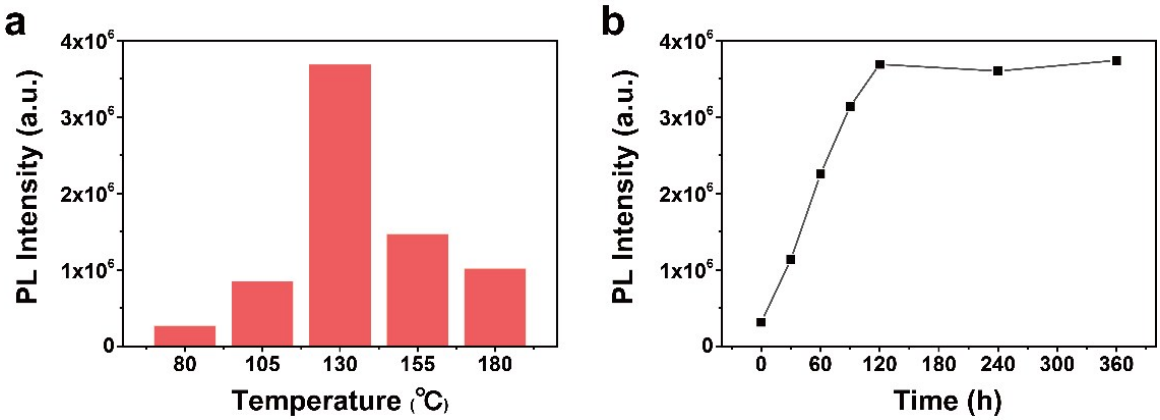


Figure S2 (a). The overall PL intensity of T-CQDs measured under different reacting temperature. (b). the variation of overall PL intensity of T-CQDs measured after different reaction time.

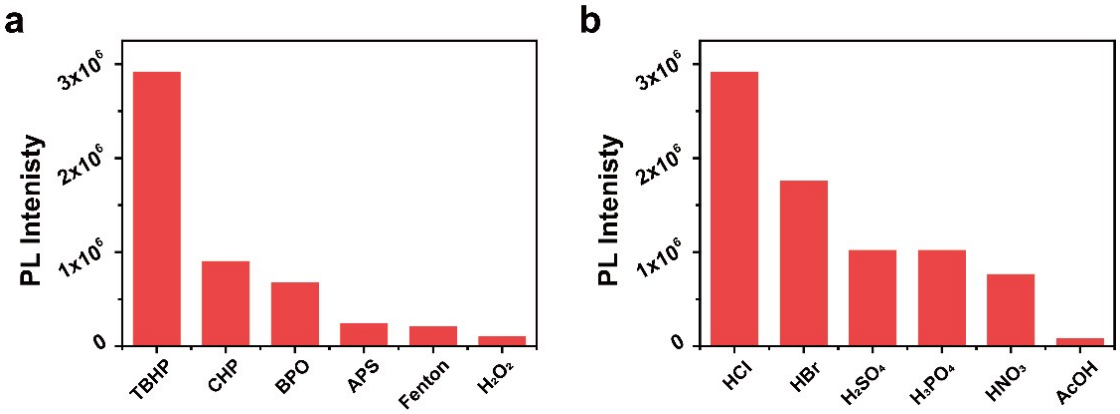


Figure S3 (a). The overall PL intensity of T-CQDs synthesized with different oxidative radical reagents. (b). The overall PL intensity of T-CQDs synthesized with different acids.

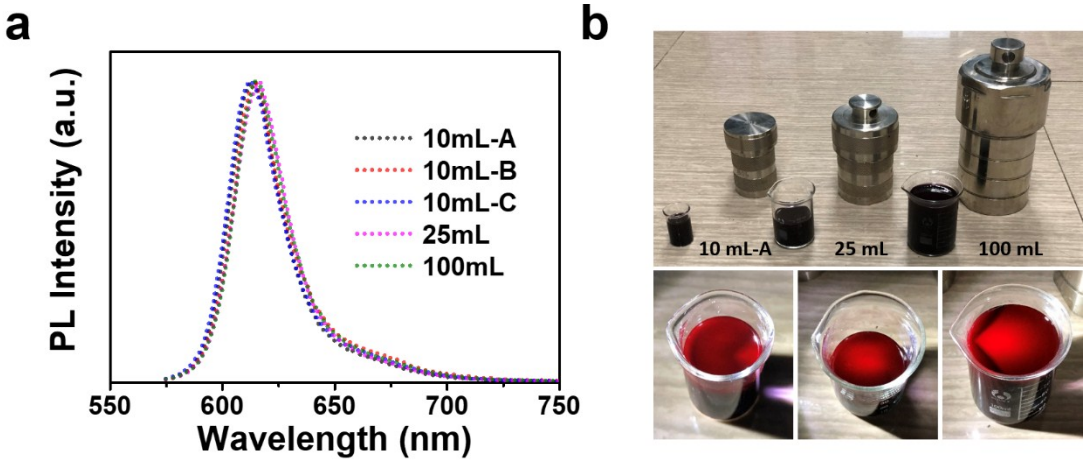


Figure S4 (a). The normalized PL emission spectra of T-CQDs synthesized in different batches. (b). the photographs of different size autoclaves and the resultant crude T-CQDs product solutions.

Table S1 Summarized optical performance and synthesis yield of the T-CQDs synthesized from 5 different batches.

Batch	λ_{em} (nm)	FWHM (nm)	PLQY	Yields
10mL-A	615	26	80.77%	17.64%
10mL-B	613	27	78.44%	16.13%
10mL-C	613	28	79.23%	17.18%
25mL	617	27	89.50%	16.73%
100mL	614	26	83.88%	18.73%
Summarize	615±2	27±1	84±5%	17.5±1.5%

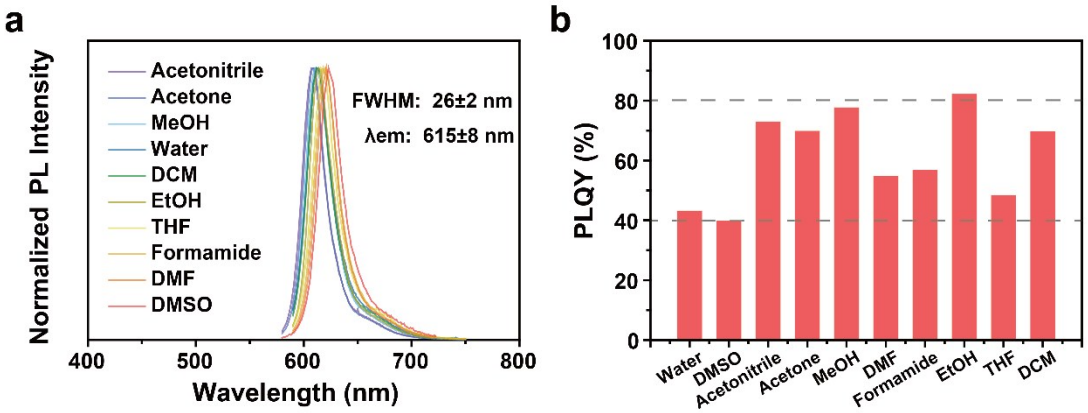


Figure S5 Summarized PL performances of T-CQDs in various solvents. (a). The normalized PL emission spectra of T-CQDs in different solvents. (b). Summarized PLQY of T-CQDs in different solvents.

Table S2 Summarized PL performances of T-CQDs in various solvents.

Solvent	λ_{em} (nm)	FWHM (nm)	PLQY (%)
Water	612	27	43.26
DMSO	623	26	39.94
Acetonitrile	608	25	73.07
Acetone	610	24	70.01
MeOH	610	28	77.82
DMF	621	24	54.93
Formamide	620	27	56.97
EtOH	614	26	82.36
THF	616	26	48.49
DCM	613	26	69.82

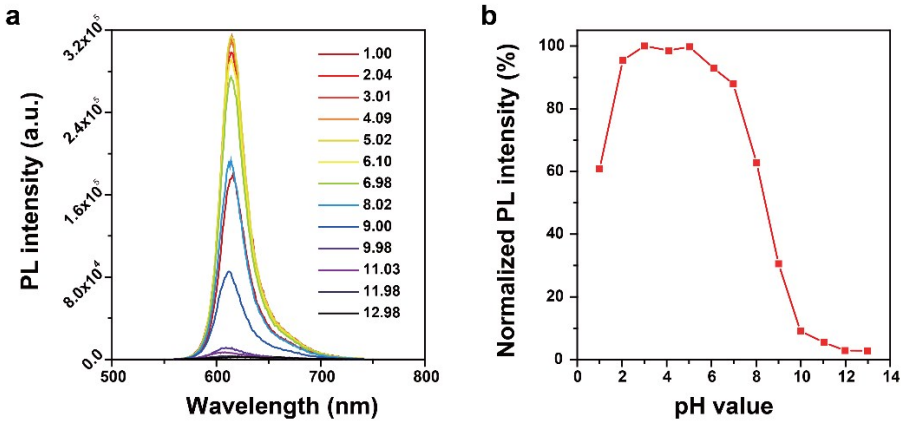


Figure S6 pH effect on the fluorescence intensity of T-CQDs (concentration: 10 μg/mL). (a) The PL spectra of T-CQDs in aqueous solutions with different pH values; (b) Normalized PL intensity of T-CQDs in aqueous solutions with different pH values.

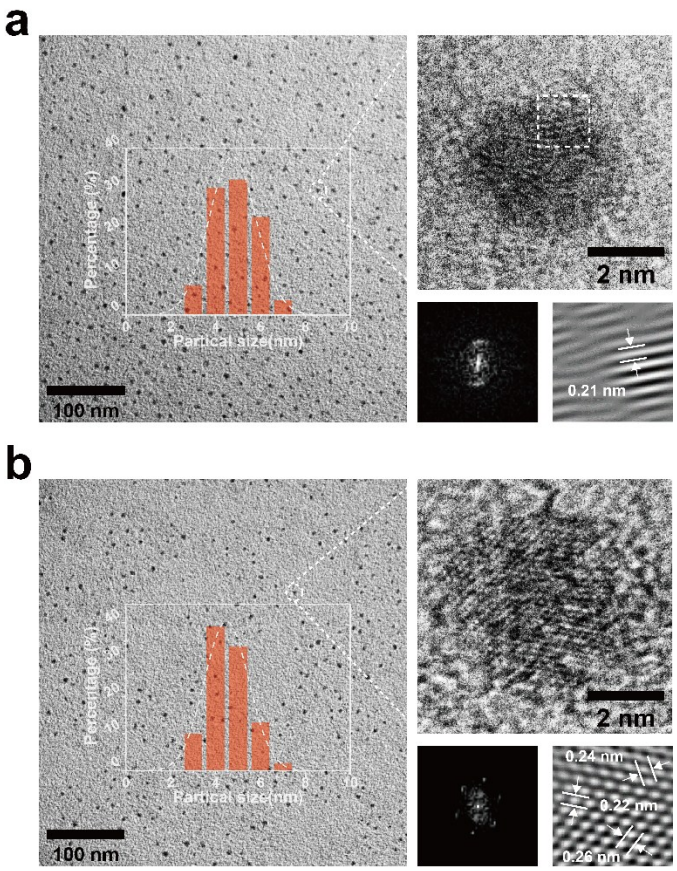


Figure S7 The TEM image of P-CQDs (a) and D-CQDs (b). Average lateral size of P-CQDs and D-CQDs were 4.75 and 4.49 nm, respectively.

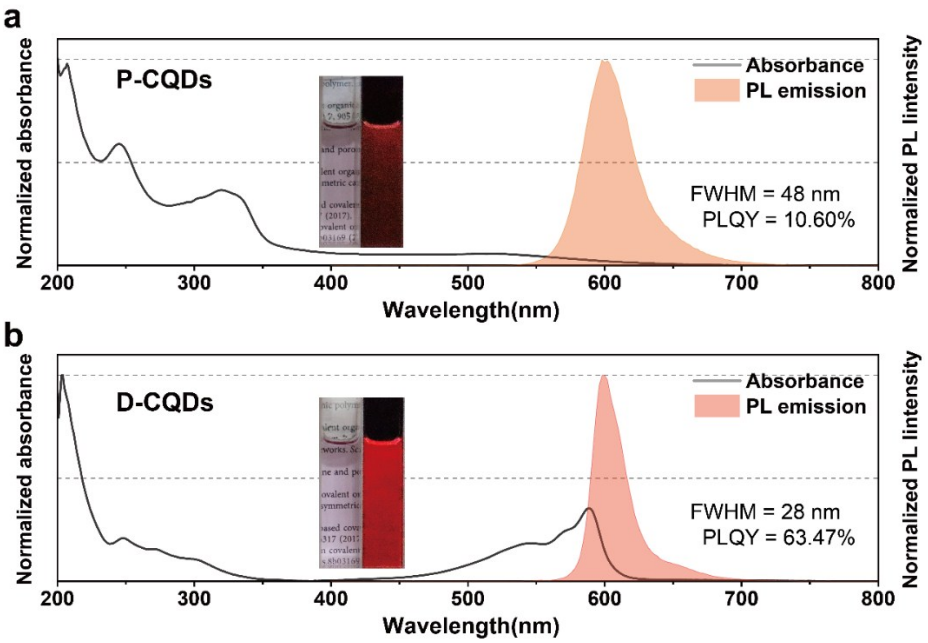


Figure S8 (a). The UV-absorbance, PL emission and photographs (inserted) of P-CQDs (10 μg/mL ethanol solution). (b). The UV-absorbance, PL emission and photographs (inserted) of D-CQDs (10 μg/mL ethanol solution).

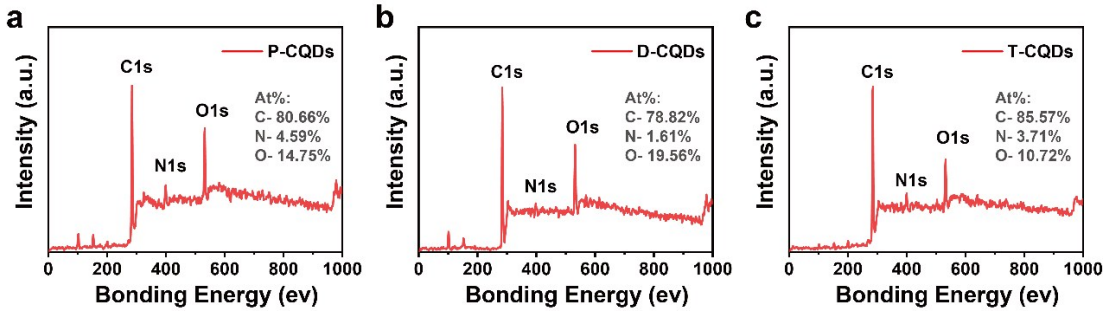


Figure S9 The XPS survey of P-CQDs (a), D-CQDs (b) and T-CQDs (c).

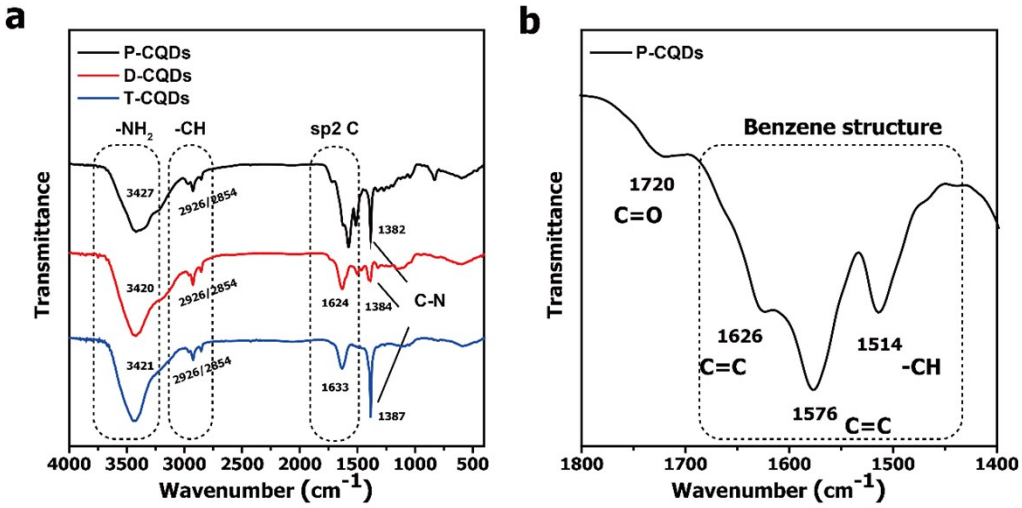


Figure S10 (a). The FT-IR spectra of P-CQDs, D-CQDs and T-CQDs; (b). The FT-IR spectrum (1400~1800 cm^{-1}) of P-CQDs.

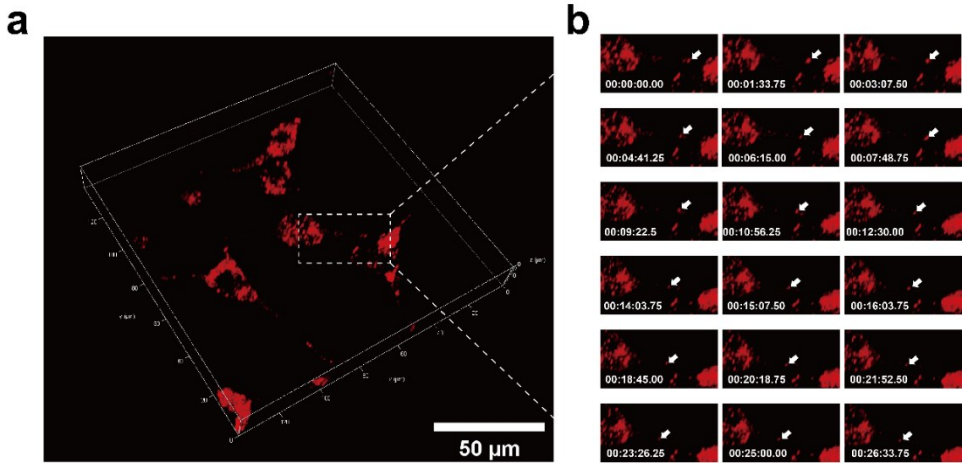


Figure S11 The time-resolved 3D imaging results of lysosomes in the living NIH-3T3 cells (a). A single frame of the 3D-reconstructed confocal image of NIH-3T3 cells. (b). time-resolved frames of the selected area for tracking individual lysosome movements. (For live video record, please see Video S1)