

Supporting Information

Nitrogen and chlorine co-doped carbon dots with synchronous excitation of multiple luminous centers for blue-white emission

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Materials

p-Phenylenediamine was purchased from Shanghai Macklin Biochemical Co., Ltd. Melamine and aluminum chloride hexahydrate were purchased from Shanghai Aladdin Biochemical Technology Co., Ltd. The silica gel and mobile phase used in Column chromatography isolation were obtained from Shanghai yuanye Bio-Technology Co., Ltd. and Shanghai Titan scientific Co., Ltd., respectively. Hydrazine hydrate was purchased from Sinopharm Chemical Reagent Co. (Shanghai, China). All reagents used in the experiments are commercially attainable without further purification. Ultrapure water was produced by Millipore system from Hunan Kertone Water Treatment Co., Ltd.

Apparatus

Morphology of the samples is analysed by transmission electron microscopy (TEM; JEM-2100). The structure and component of sample are recorded by Fourier transform infrared (FT-IR) spectroscopy (performed with Vertex80+Hyperion2000) and X-ray photoelectron spectrometer (XPS; performed with ESCALAB 250Xi). The photoluminescence (PL) and ultraviolet-visible (UV-vis) spectrum of the samples are observed by a fluorescence spectrophotometer (F-4500) and an UV-1800PC spectrophotometer, respectively. The absolute optical quantum yield (QY) is measured by the Horiba FLSP920 system with a calibrated integrating sphere. The Raman spectra of the samples is obtained by a NEXUS-870 spectrometer with a 532 nm laser.

Synthesis of melem hydrazine (*2,5,8-trihydrazino-s-heptazine*)

1.6 g (7.5 mmol) of melem was dispersed in 15mL (0.25 mol) of 55% aqueous hydrazine hydrate solution and sealed in a 40 mL Teflon lined autoclave. The autoclave was heated at 140°C in an oven for 12 h hours. The obtained yellowish suspension liquid was moved to a beaker and 10% aqueous solution of HCl was added to adjust pH between 1–2. This solution was centrifuge to wipe off un-reacted solid residue. The obtained solution was precipitated by adding 10% aqueous solution of NaOH to adjust pH between 7.5 – 8.5. The obtained solid was again dissolved in aqueous solution of HCl, centrifuged and re-precipitated in aqueous solution of NaOH and this procedure was repeated for three times. Ultimately, obtained solid was rinsed several times with DI water and ethanol then dried under vacuum. All the characterization data of acquisition were consistent with the previously reported literature (Figure S1).³²

Synthesis of N,Cl-CDs

84 mg of p-phenylenediamine (pPD) was dissolved in 5 mL ethanol. 17 mg melem hydrazine (MH) was dispersed in 30 mL deionized water. Mix them up and got a suspension, then add 16 mg aluminium chloride hexahydrate as a catalyst for the aromatic electrophilic addition reaction. Put them in a 40 mL Teflon lined autoclave after intensive mixing. The autoclave was heated at 180°C in an oven for 10 h.

Column chromatography isolation

We separated the as-prepared sample by column chromatography on silica gel. Petroleum

ether, ethyl acetate and methanol were used as mobile phase. The mobile phase consists of petroleum ether and ethyl acetate. Percentage composition of ethyl acetate gradually increased from 0 to 100. When mobile phase was purified ethyl acetate, we substituted methanol for petroleum ether. Percentage composition of methanol gradually increased from 0 to 100. N,Ci-CDs, showed orange fluorescence in the column under ultraviolet light, can be collected when the mobile phase was purified ethyl acetate.

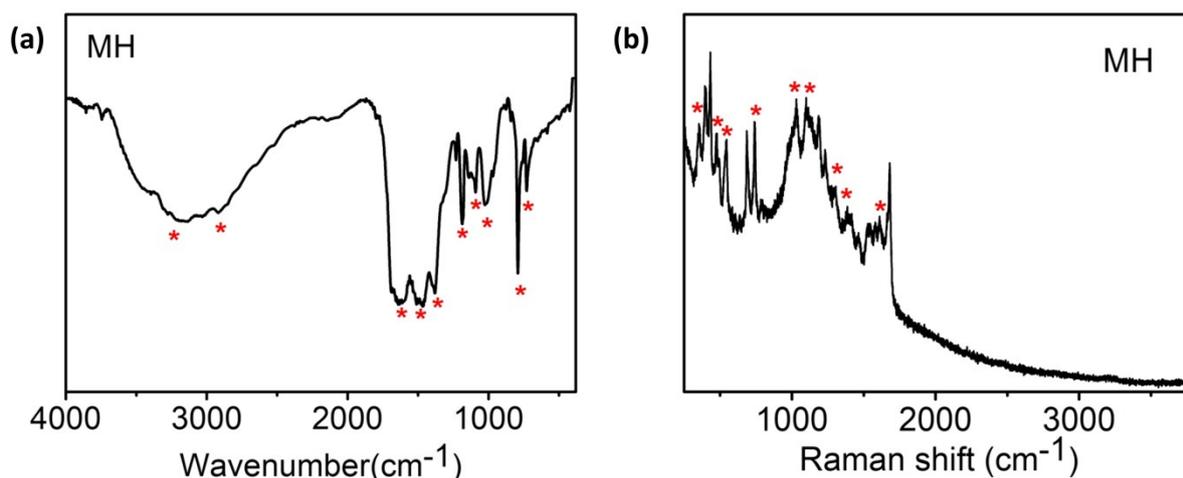


Figure S1. (a) FT-IR spectrum of MH. (b) Raman spectrum of the MH.

FTIR: 3203, 2912, 1608, 1467, 1382, 1186, 1096, 1011, 790, 725 cm^{-1} .

Raman: 1608, 1382, 1307, 1097, 1031, 741, 543, 471, 350 cm^{-1} .

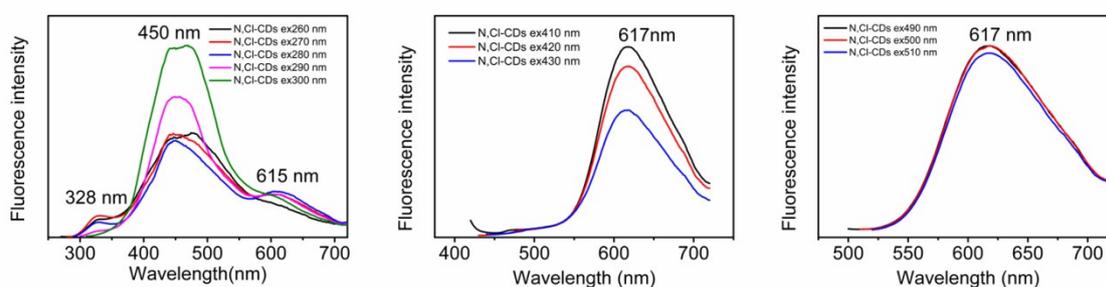


Figure S2. PL emission spectra of N,Ci-CDs under different excitation wavelengths (a) 260-300 nm, (b) 410-430 nm and (c) 490-510 nm.

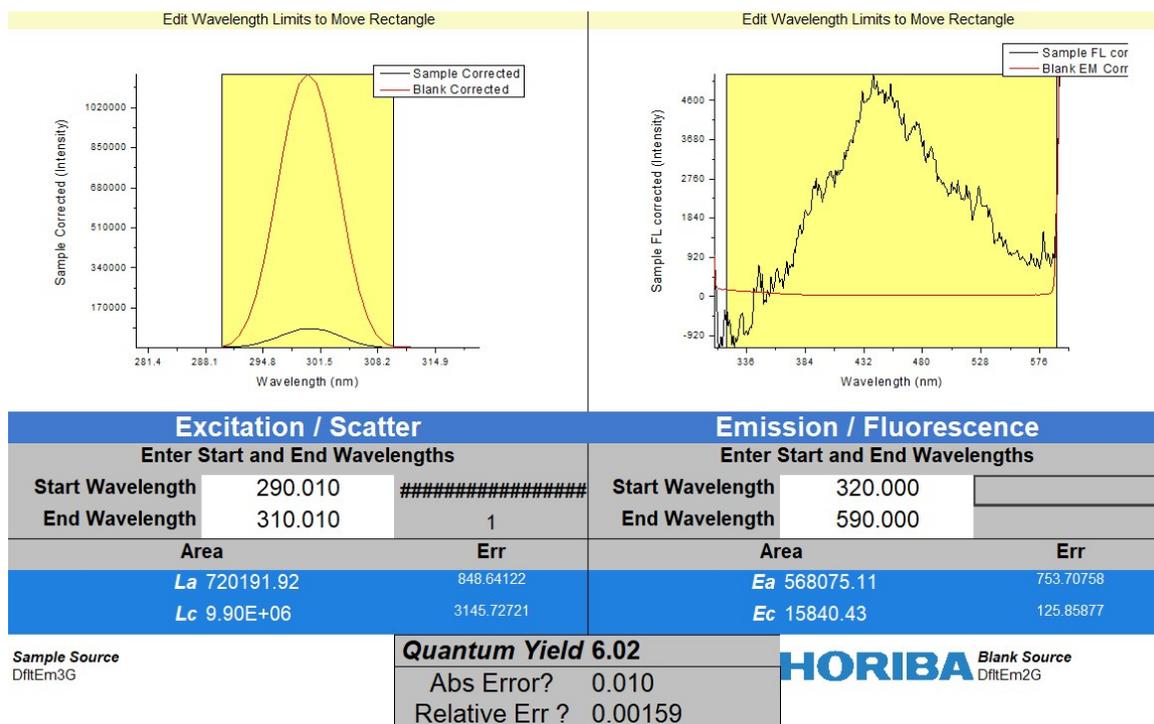


Figure S3. The absolute fluorescence quantum yield of N,Cl-CDs.

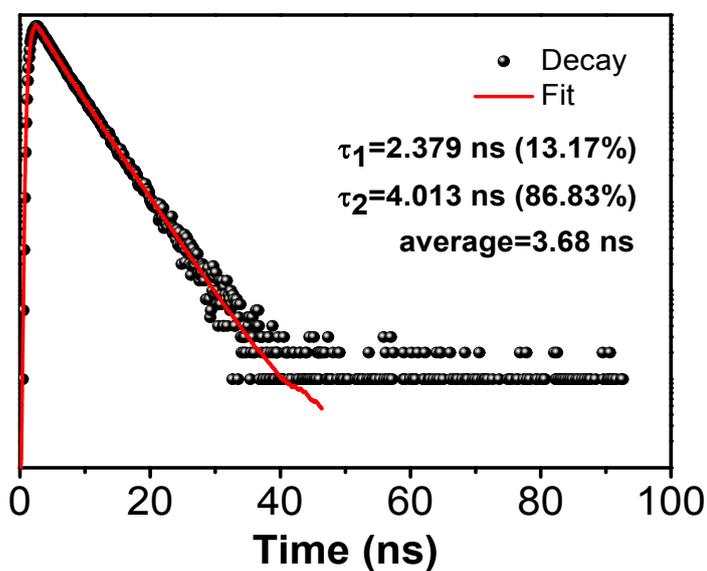


Figure S4. Time-resolved PL spectrum of the N,Cl-CDs.

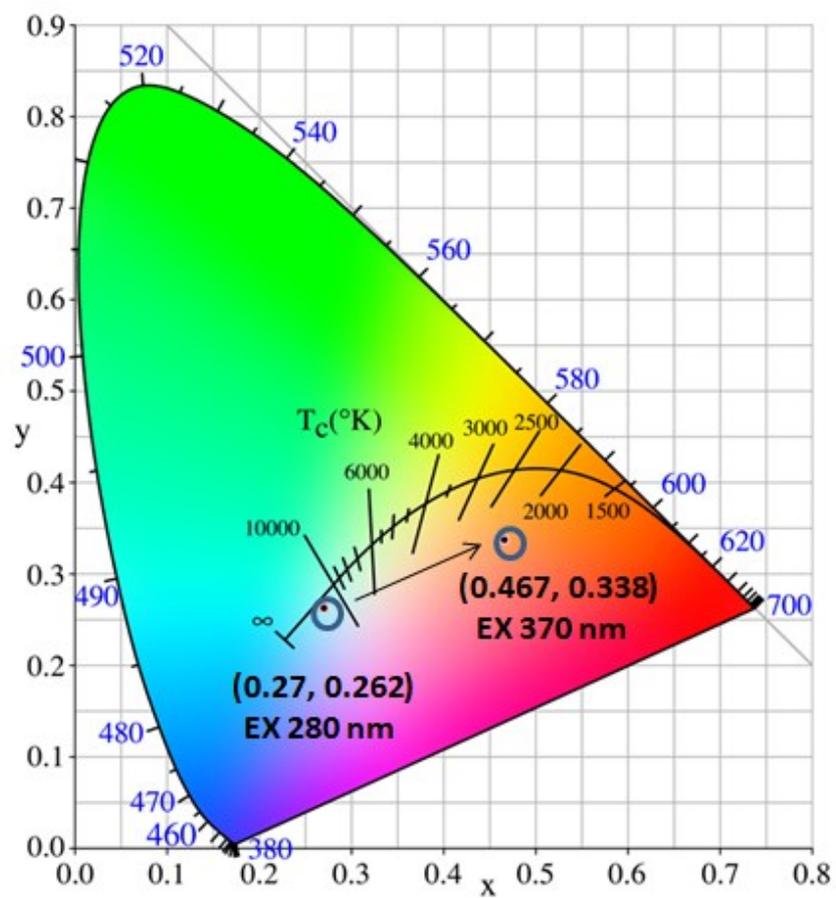


Figure S5. CIE color coordinate of the N,Cl-CDs.