Supporting Information

Achieving High-Stability Aqueous Room-Temperature Phosphorescent Materials via In Situ Host-Guest Strategy

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Experimental Sections

1.1 Materials

Cyanuric acid (CA), ethylenediamine and cysteine was purchased from Macklin Chemicals Co. Ltd (Shanghai, China). Diethylenetriamine, ethanolamine was purchased from Aladdin Chemicals Co. Ltd (Shanghai, China). Phosphoric acid was purchased from Sinopharm Chemical Reagent Co. Ltd (Shanghai, China). The pomelo was purchased from local supermarket and washed with water for further use. All chemicals were used as received without further purification unless otherwise specified. Deionized water was used throughout this study.

1.2 Measurements

The morphologies of NS-CDs_{20}@CA were examined using a JEOL JEM 2100 Transmission Electron Microscope (TEM). X-ray diffraction (XRD) analyses were performed using a X'pertpro X-ray diffractometer (Netherlands). Field emission scanning electron microscopy (SEM) was operated on a Hitachi S-4800 microscope. Optical absorption spectra were recorded on a Shimadzu UV-3600 spectrophotometer. Photographs were captured under UV lamp illumination at 365 nm using a REDMI K30. Fourier Transform Infrared Spectroscopy (FTIR) spectra were acquired using a Thermo Fisher Scientific Nicolet iS5 spectrometer, while the X-ray photoelectron spectroscopy (XPS) spectra were obtained with a Thermo Science K-Alpha. The prompt/delayed PL spectra were measured using an Edinburg FLS1000 fluorescence spectrophotometer (Edinburgh Instruments, UK). The absolute total photoluminescent quantum yields were measured using an integral sphere on FLS 1000 equipment, which encompasses both fluorescent and afterglow emission components. The phosphorescent quantum yield (QY) was determined through peak-differentiation-imitating analysis,[1, 2] wherein the phosphorescent and fluorescent quantum yields can be derived from their respective integrate area ratios identified in the steady-state photoluminescence (PL) spectrum using the following equation:

$$Phosphorescent\ QY = \Phi\frac{A_2}{A_1 + A_2} \quad (1)$$

where A_1 and A_2 are the integral areas of fluorescence and phosphorescence from the steady-state PL spectrum, respectively.

The multiexponential PL decays [I(t)] of X-CDs$_{20}$@CA were described using the following equation:

$$I(t) = \sum_i B_i e^{-\frac{t}{\tau_i}} \quad (2)$$

where B_i and τ_i represent the amplitudes and lifetimes of the individual components for multiexponential decay profiles, respectively.

1.3 Synthesis of materials

1.3.1 Preparation of NS-CDs$_{20}$@CA

Initially, 30 mL of deionized water, 2.58 g of cyanuric acid (20 mmol), 1.21 g of cysteine (10 mmol), and 0.6 mL of EDA (10 mmol) were sequentially added to a poly(tetrafluoroethylene) (Teflon)-lined autoclave (50 mL) and stirred for 5 minutes to ensure thorough mixing. The autoclave was then heated in an oven at 180 °C for 13 h and allowed to cool naturally to room temperature. Subsequently, the reaction product was centrifuged multiple times with deionized water to remove impurities and vacuum-dried to obtain the solid powder NS-CDs$_{20}$@CA. The synthesis procedure for the other X-CDs$_{20}$@CA samples was identical to the one described above, with the only variation being the change in reactant precursors.

1.3.2 Preparation of 1-CDs

1-CDs were prepared by hydrothermal treatment of pomelo peel. In a typical synthesis, 1 g of pomelo peel was added into 15 mL of H_2O. Then the mixture was transferred into a 25-mL Teflon-lined autoclave and heated at 200 °C for a period of 3 h. The 1-

CDs were collected by removing the large dots through centrifugation at 12000 rpm for 10 min and finally dried under vacuum for 48 h.[3]

1.3.3 Preparation of 2-CDs

In a typical process, diethylenetriamine (3.0 mL) and phosphoric acid (1.0 mL) were dissolved in deionized water (20 mL) to form a clear dispersion. The resultant dispersion was transferred to a Teflon-inlet stainless steel autoclave (50 mL), heated at 200 ºC for 5.0 h, and then naturally cooled down to room temperature. Finally, the 2-CDs were obtained by dialyzing with a dialysis membrane (MWCO: 500 Da) for 2 days and heat drying, respectively.[4]

1.3.4 Preparation of 3-CDs

Ethanolamine (4.0 mL) was dissolved in deionized water (16 mL), and 8.0 mL of phosphoric acid was added dropwise while stirring. After stirring for 30 min, the mixture formed was transferred to a 100 mL beaker, and heated in a domestic oven for 4 min (700 W). After being cooled to room temperature, the obtained brownish yellow gel-like solid was dissolved by adding deionized water (30 mL). The crude product was centrifuged at 10000 rpm for 15 min to remove large particles, and then pH value of the solution was adjusted to neutrality with sodium carbonate. After a filtration against 0.22 μm membrane, the resulting filtrate was dialyzed via a dialysis membrane (1000 MWCO) for one week. After a freeze-drying, the final product i.e. pale yellow 3-CDs powder was harvested.[5]

Results and Discussion

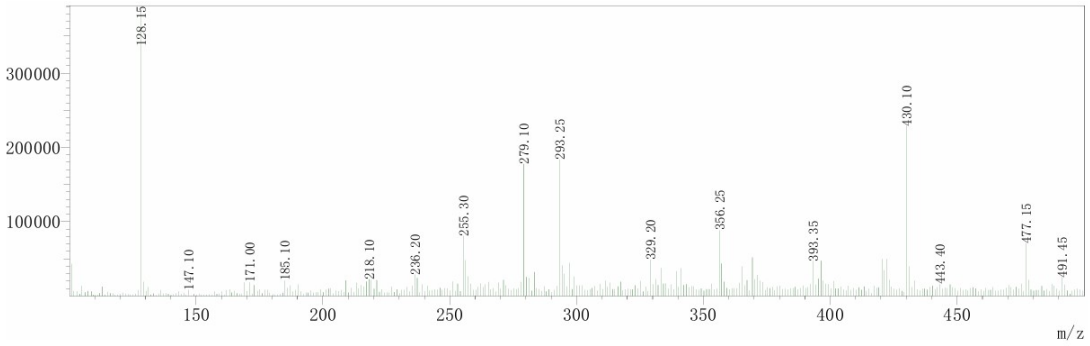

Fig. S1 Mass spectrometry of NS-CDs$_{20}$@CA.

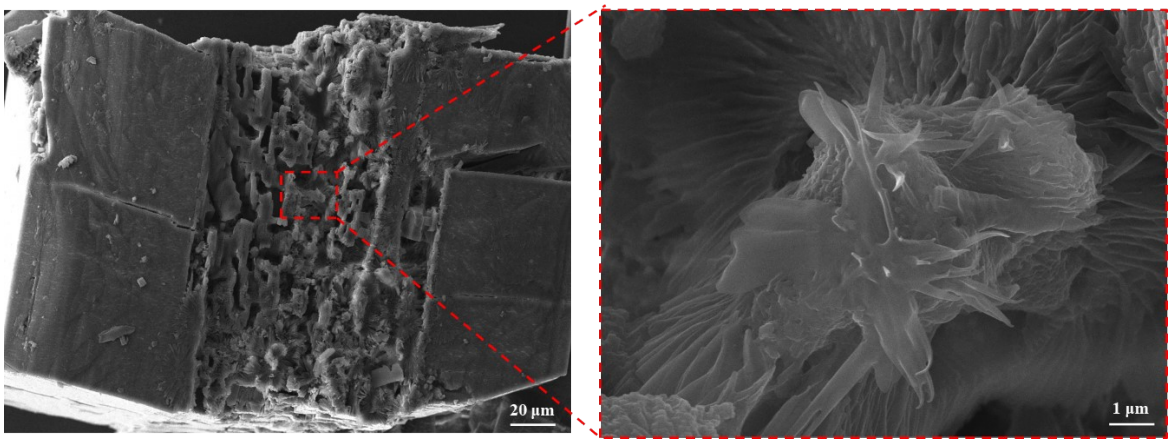

Fig. S2 SEM images of NS-CDs$_{20}$@CA.

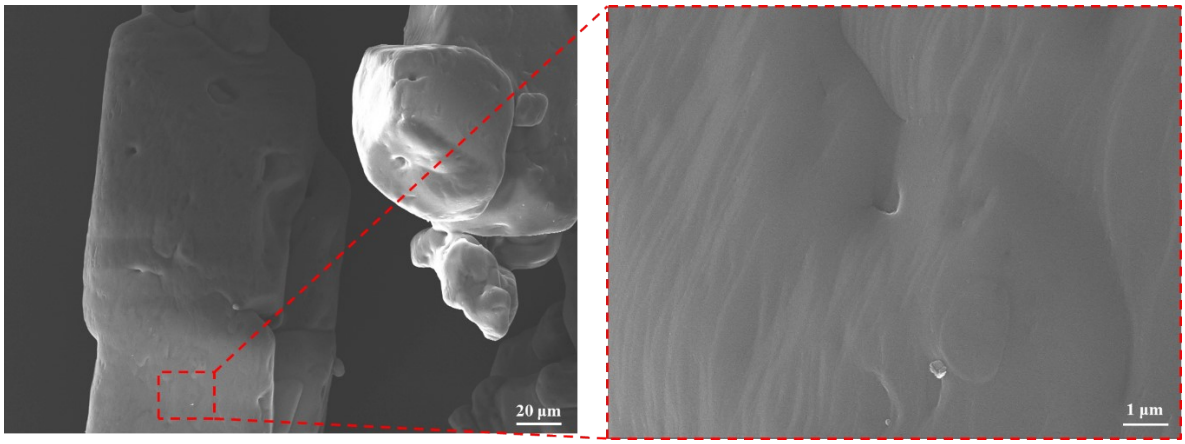

Fig. S3 SEM images of CA.

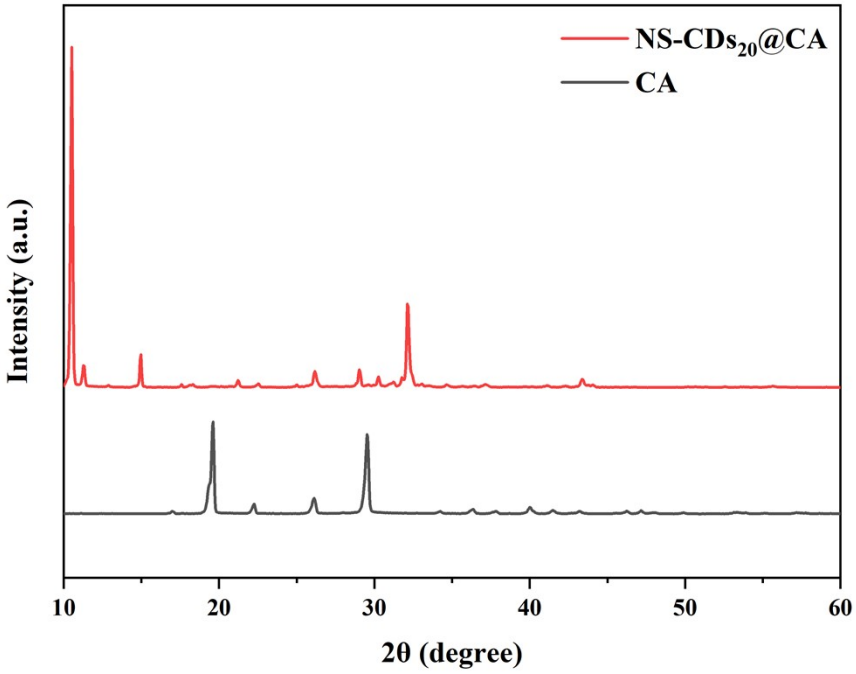

Fig. S4 The XRD curves of CA and NS-CDs$_{20}$@CA, respectively.

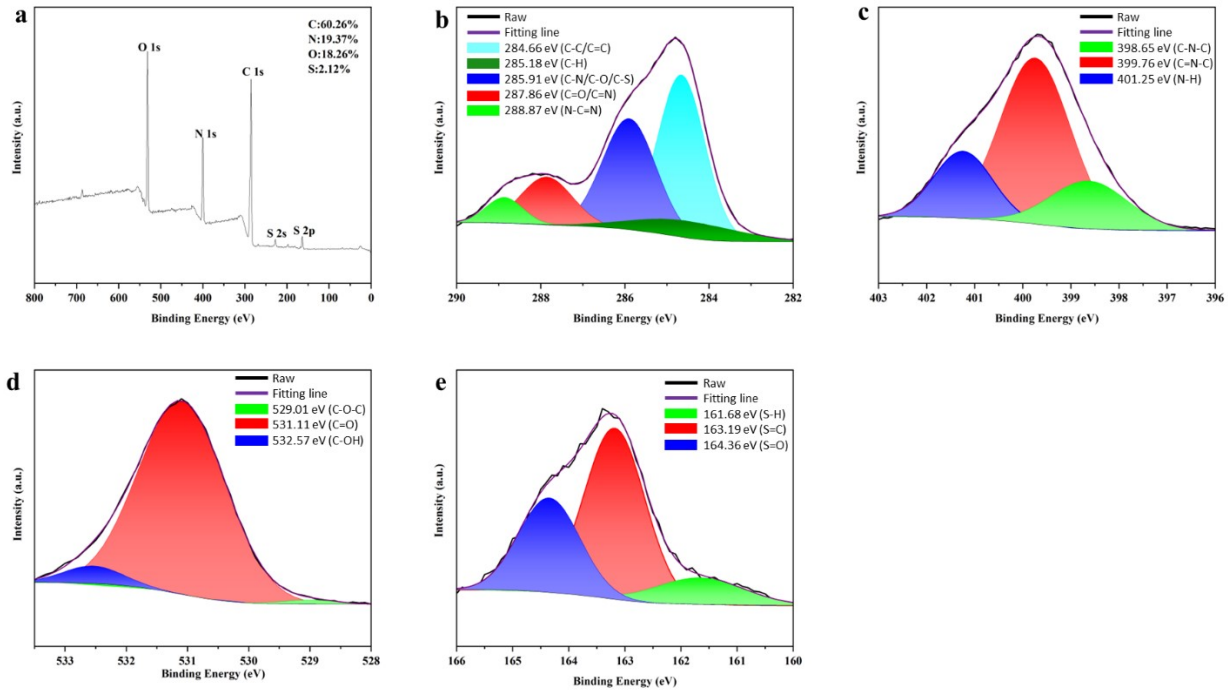

Fig. S5 XPS survey (a), high-resolution C 1s (b), N 1s (c) O 1s (d) and S 2p (e) spectra of NS-CDs$_1$, respectively.

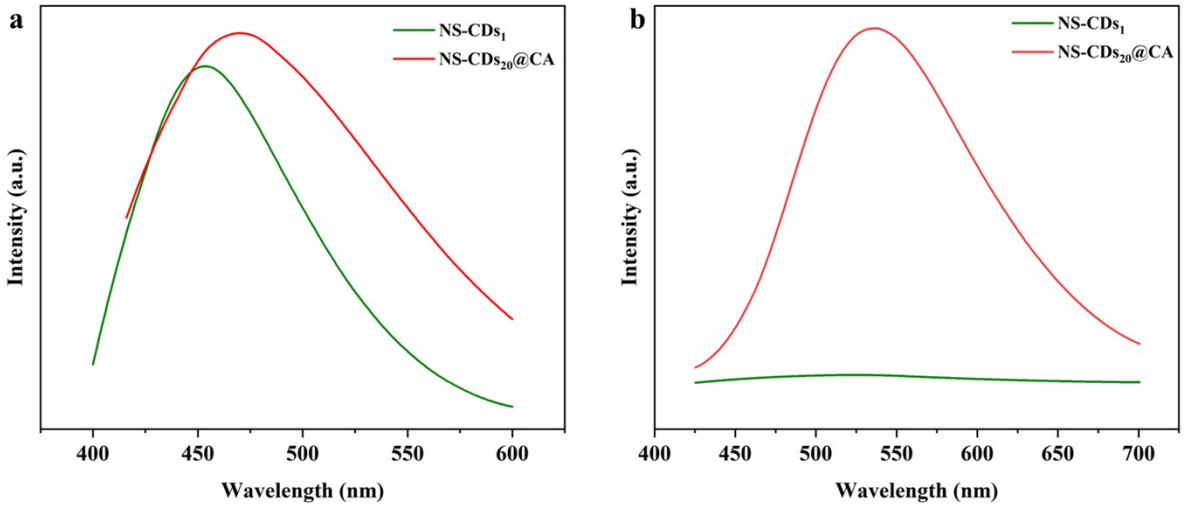

Fig. S6 Fluorescence (a) and Phosphorescence (b) spectra of NS-CDs_1 and NS-CDs_{20}@CA, respectively.

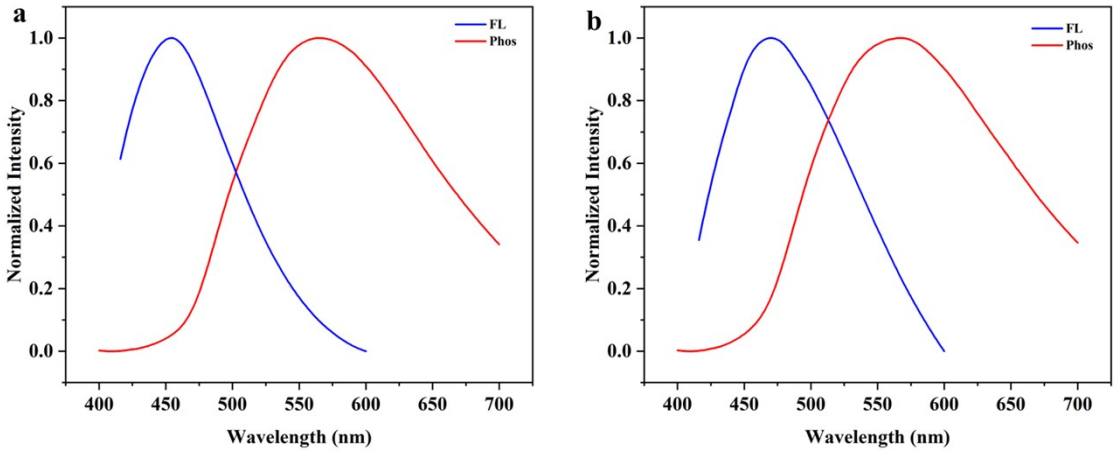

Fig. S7 Fluorescence and Phosphorescence spectra of N-CDs_{20}@CA (a) and NS-CDs_{20}@CA (b) measured at 77K, respectively.

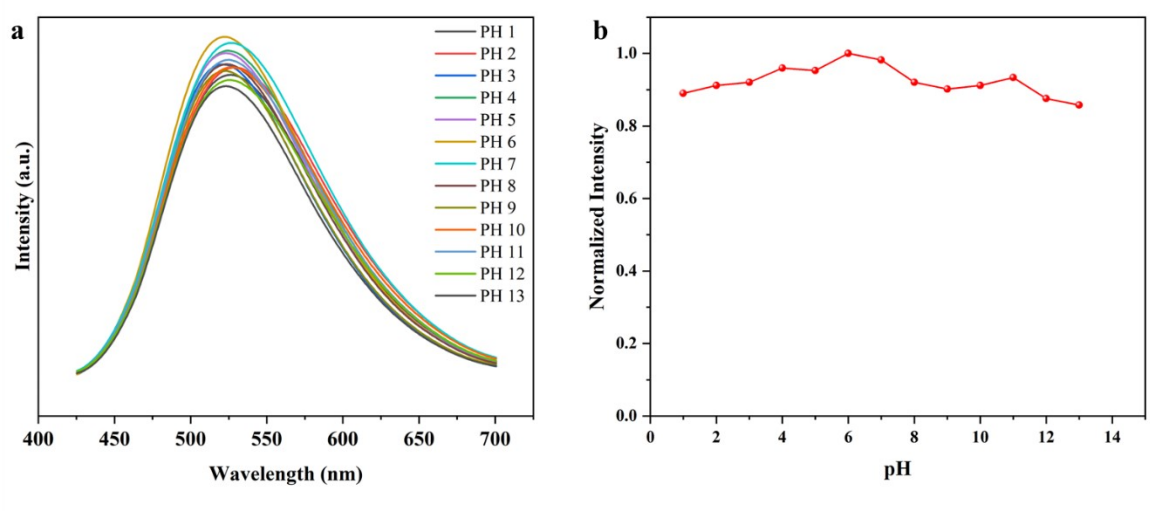

Fig. S8 Phosphorescent spectra (a) and normalized intensities (b) of NS-CDs_{20}@CA with different pH values under ambient conditions.

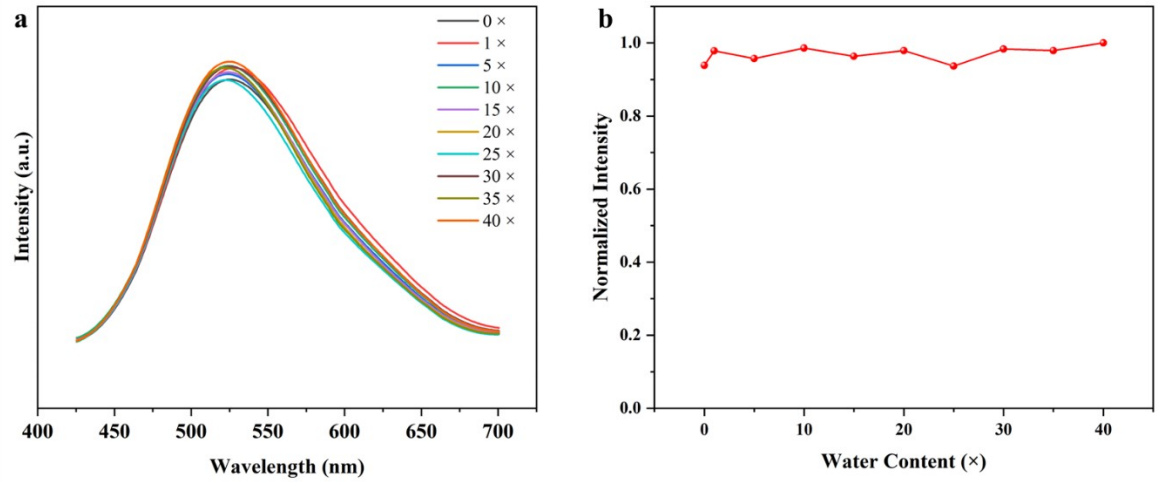

Fig. S9 Phosphorescent spectra (a) and normalized intensities (b) of NS-CDs_{20}@CA for different water content.

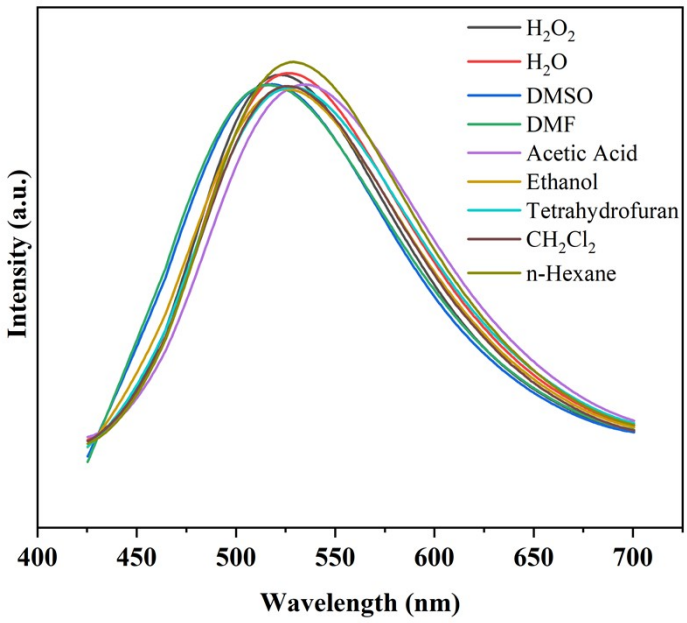

Fig. S10 Phosphorescent spectra of NS-CDs_{20}@CA with incorporating of various solvents.

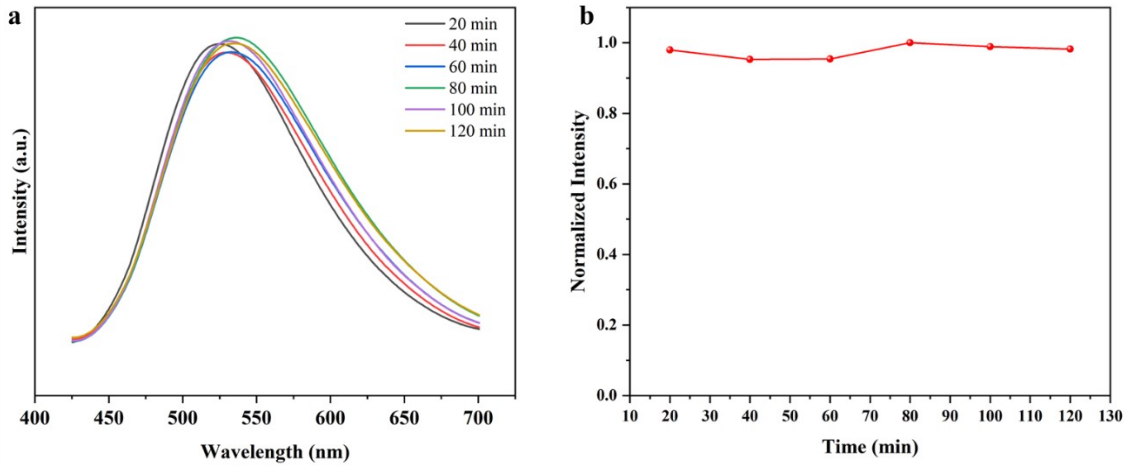

Fig. S11 Phosphorescent spectra (a) and normalized intensities (b) of NS-CDs_{20}@CA versus time (20 minutes intervals) under illumination of UV light for 120 minutes.

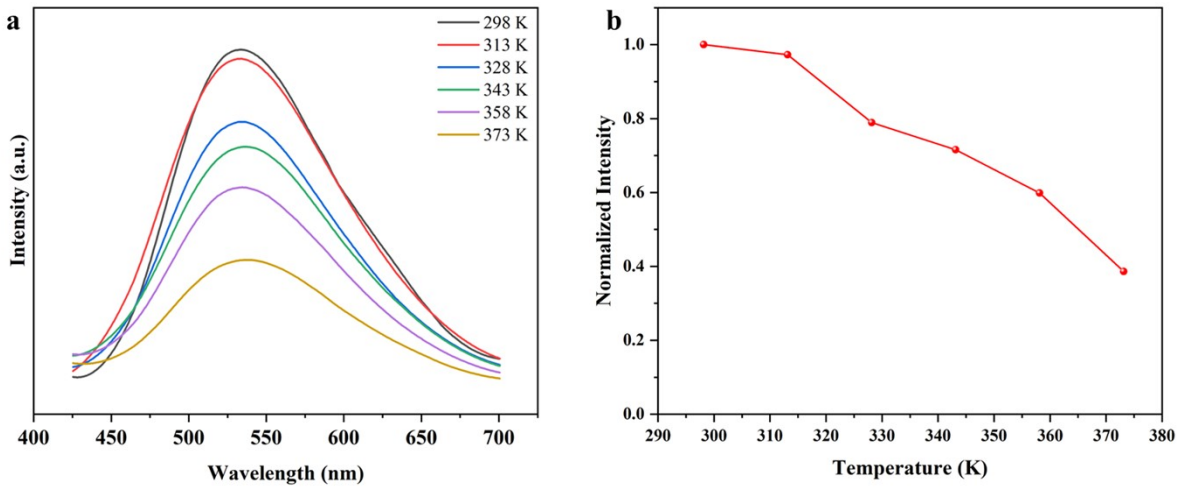

Fig. S12 Phosphorescent spectra (a) and normalized intensities (b) of NS-CDs_{20}@CA at different temperatures from 298 K to 373 K.

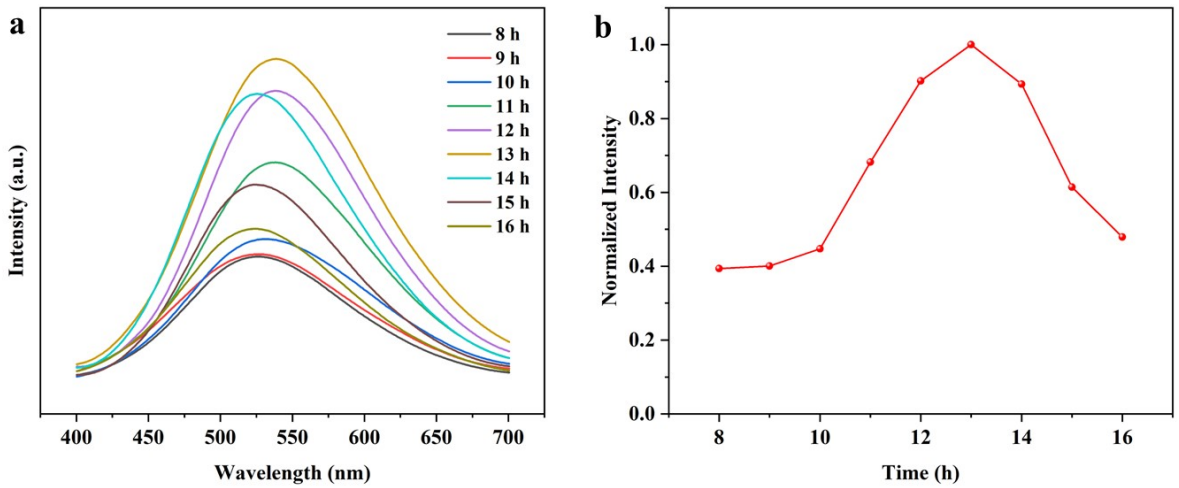

Fig. S13 Phosphorescent spectra (a) and normalized intensities (b) of NS-CDs_{20}@CA at 180 ℃ for varying durations of reaction.

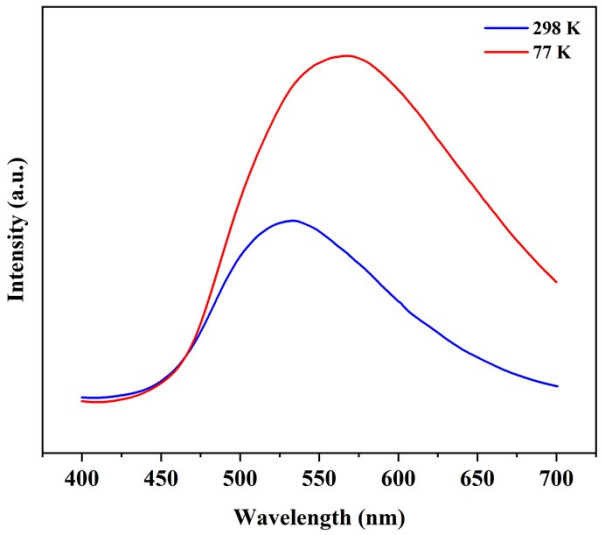

Fig. S14 Phosphorescence spectra of NS-CDs$_{20}$@CA measured at 77K and 298 K.

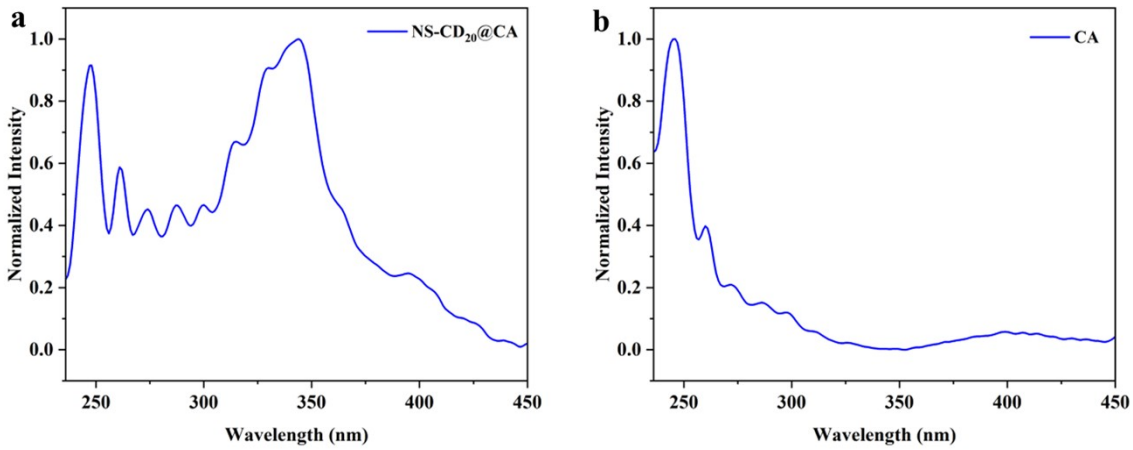

Fig. S15 Phosphorescence excitation spectra of NS-CDs$_{20}$@CA (a) and CA (b).

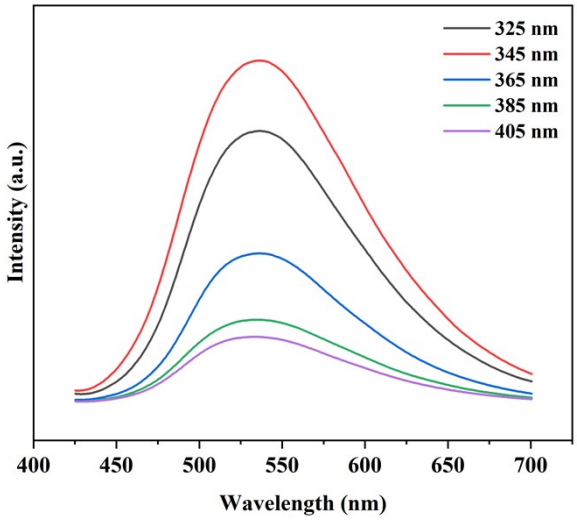

Fig. S16 Phosphorescence spectra of NS-CDs_{20}@CA at different excitation wavelength from 325 nm to 405 nm.

Table S1 Fitting parameters of the RTP decay curves of CDs_{20}@CA, N-CDs_{20}@CA and NS-CDs_{20}@CA, respectively.

Sample	τ_1 (ms)	B_1 (%)	τ_2 (ms)	B_2 (%)	τ_3 (ms)	B_3 (%)	τ_{avg} (ms)
CDs_{20}@CA	0.28	8.12	1.97	33.45	18.81	58.43	11.68
N-CDs_{20}@CA	17.88	2.12	185.44	32.74	763.92	65.14	558.72
NS-CDs_{20}@CA	49.83	4.87	250.44	50.57	930.15	44.56	543.53

Table S2 The PLQYs and PhQYs of CDs_{20}@CA, N-CDs_{20}@CA and NS-CDs_{20}@CA, respectively.

Sample	Φ_{PLQY} (%)	Φ_{PhQY} (%)
CDs_{20}@CA	0.59	0.0053
N-CDs_{20}@CA	31	7.44
NS-CDs_{20}@CA	23.36	9.73

Supplementary References

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