Supplementary Information for

White Emissive Carbon Dots Actuated by the H- / J-Aggregates and Förster Resonance Energy Transfer

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Materials and Methods

1. Synthesis of WCDs.

Healthy white light emitting CDs was synthesized by using catechol and o-phenylenediamine (PDA) as the precursor. In a typical experiment, 1) dissolved the precursor in the DMF-NAA/NAA mixed solvent by ultrasonic wave for 30 minutes, the solution color should be bright red, 2) put it in a high temperature high pressure autoclave (20 mL), then placed inside a high temperature (180 ^{o}C) drying oven without any greenhouse warming for 12 hours. The resultant liquid was then washed with petroleum ether several times to remove all unreacted and potentially detrimental surface species, the centrifugation speed was 9000 rpm for 30 min. The products were denoted as white light emitting CDs.

2. Synthesis of CD-OH.

Carbon dots (CD-OH) was synthesized by using catechol as the precursor. In a typical experiment, 1) dissolved the precursor in the DMF-NAA/NAA mixed solvent by ultrasonic wave for 30 minutes, the solution should be colorless, 2) put it in a high temperature (180 ^{o}C) high pressure autoclave (20 mL), then placed inside a high temperature drying oven without any greenhouse warming for 12 hours. The resultant liquid was then washed with petroleum ether several times to remove all unreacted and potentially detrimental surface species, the centrifugation speed was 9000 rpm for 30 min.

3. Synthesis of CD-NH_2.

Carbon dots (CD-NH_2) was synthesized by using o-phenylenediamine (PDA)

as the precursor. In a typical experiment, 1) dissolved the precursor in the DMF-NAA/NAA mixed solvent by ultrasonic wave for 30 minutes, the solution color should be bright light yellow, 2) put it in a high temperature high pressure autoclave (20 mL), then placed inside a high temperature (180 ^{o}C) drying oven without any greenhouse warming for 12 hours. The resultant liquid was then washed with petroleum ether several times to remove all unreacted and potentially detrimental surface species, the centrifugation speed was 9000 rpm for 30 min.

4. Preparation of the CDs/silicone resin Composites.

A certain amount of CDs were dissolved in the silicone resin. The mixture was stirred to form a homogeneous mixture.The mixture was then put into the mold and maintained for 4 h at room temperature for curing. After 4 h curing, the white emissive white light emitting CDs/silicone resin composite was obtained.

5. Preparation of the LED Devices.

The mixture of CDs and epoxy was drop-casted on the LED chip carefully, and cured at room temperaturefor 4 h. For the WLED, 2.5 mg white light emitting CDs was mixed with 1 mL silicone resin, and then the homogeneous mixture was drop-casted on the LED chip (365 nm).

Characterizations

TEM, High-resolution TEM (HRTEM) images were recorded with an FEI-TECNAI G2 transmission electron microscope operating at 200 kV. X-Ray photoelectron spectra(XPS) were obtained on a Thermo Scientific ESCALAB 250 Multitechnique Surface Analysis. Raman spectra were collected on Lab RAM HR Raman microscope. Fluorescence emission spectra were recorded on an LS-55 fluorophotometer. Absolute quantum yield measurements were performed with a calibrated integrating sphere on an Edinburg FLS 920 spectrometer. Ultraviolet-visible absorption spectrawere recorded using Shimadzu UV-2600 spectrophotometer. PL spectra were collected on the HITACHI F-7000 fluorescence Spectrophotometer. The crystalline structure was measured by using a Bruker AXS D8 Focus X-ray diffractometer, using Cu Kα radiation (λ = 1.54056 Å).

Table S1 The elemental analysis of white light emitting CDs.

	C [%]	N [%]	H [%]
WCDs	66	14	0.15

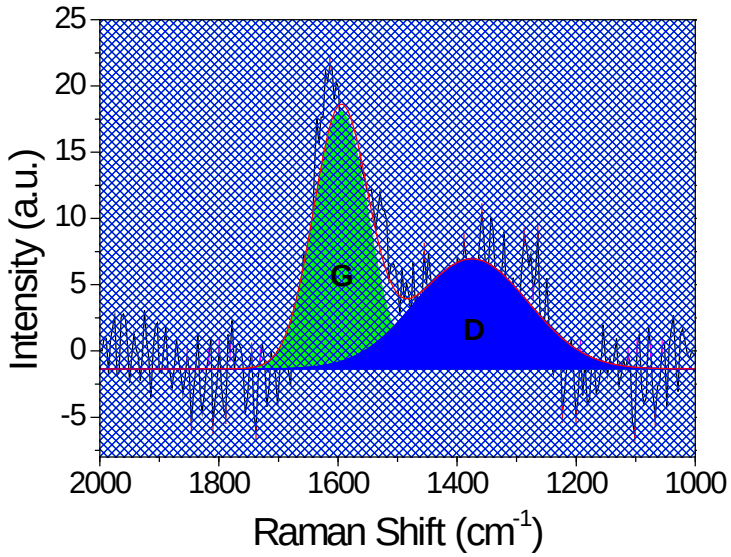


Figure S1 Raman spectrum of white light emitting CDs.

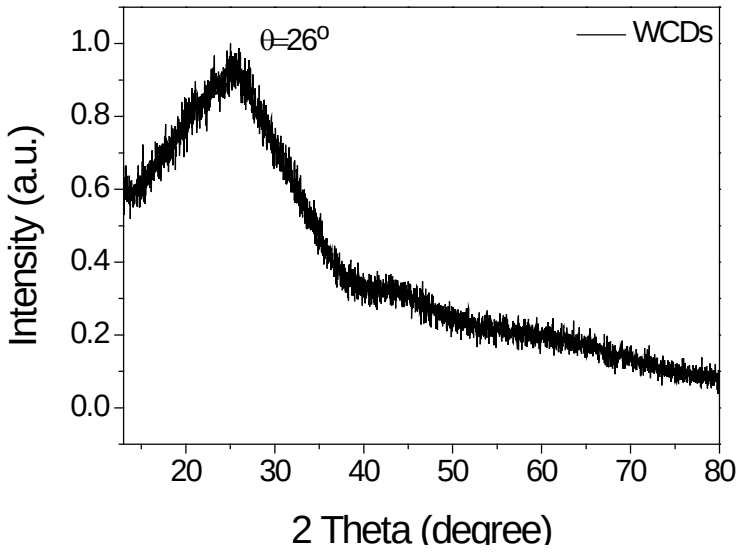


Figure S2 XRD pattern of white light emitting CDs.

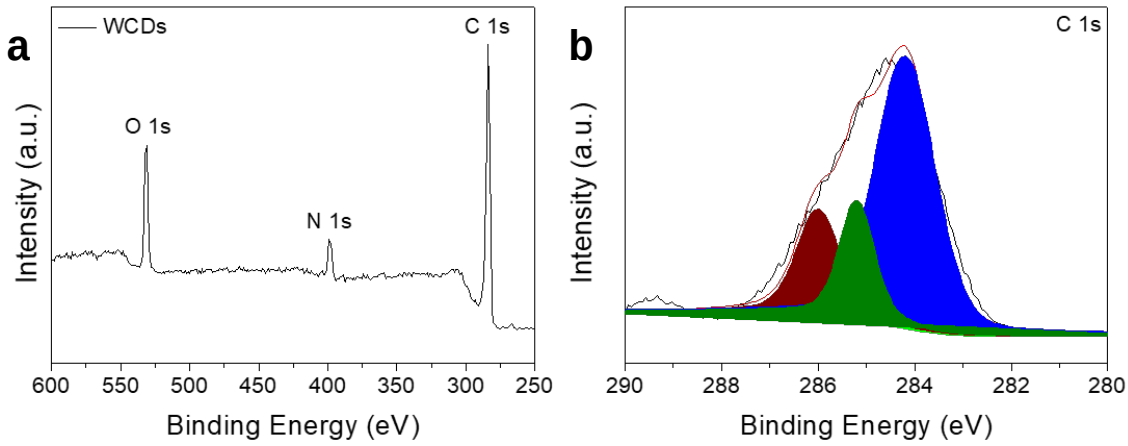


Figure S3 (a) XPS spectrum of white light emitting CDs, (b) The high-resolution C1s XPS spectrum of white light emitting CDs.

Table S2 The photoluminescent (PL) quantum yield (QY) of white light emitting CDs under different excitation wavelengths.

WCDs	Blue	Green	Red
Em [nm]	450	520	600
QY[%]	42	55	67

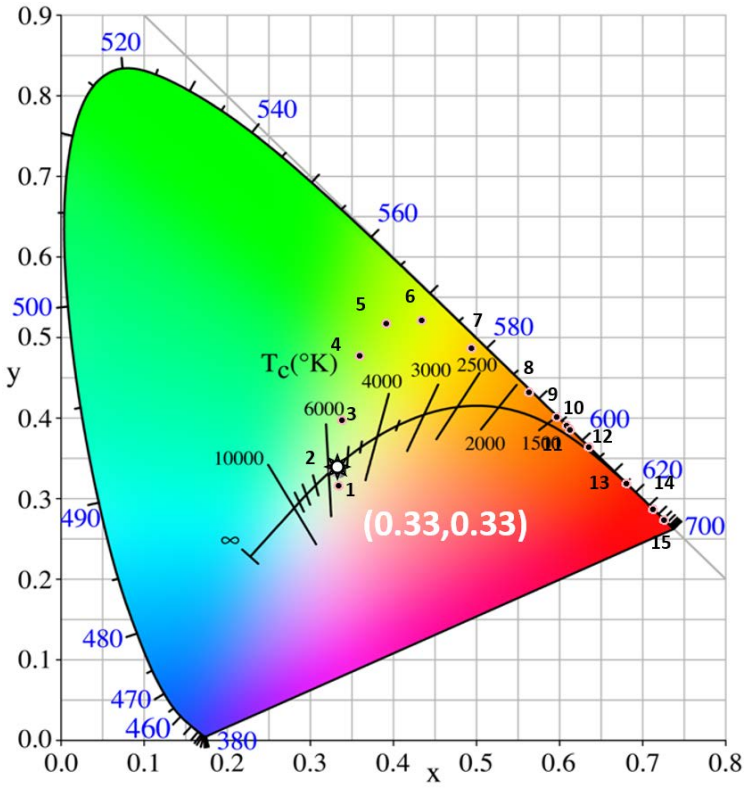


Figure S4 CIE 1931 coordinates of white light emitting CDs with different excitations (the points No. 1 and 3-15 mean the different excitation wavelengths (360-620 nm) with the interval length ~ 20 nm, the point No.2 (0.33,0.33) is excited at 365 nm).

Table S3 The CIE color coordinates of white light emitting CDs with different excitations (the points No. 1 and 3-15 mean the different excitation wavelengths (360-620 nm) with the interval length ~20 nm, the point No.2 (0.33,0.33) is excited at 365 nm).

No.	Ex [nm]	x	y
1	360	0.34	0.35
2	365	0.33	0.33
3	380	0.34	0.40
4	400	0.36	0.48
5	420	0.39	0.52
6	440	0.44	0.52
7	460	0.49	0.49
8	480	0.56	0.43
9	500	0.60	0.40
10	520	0.61	0.39
11	540	0.61	0.39
12	560	0.64	0.36
13	580	0.68	0.32
14	600	0.71	0.29
15	620	0.73	0.27

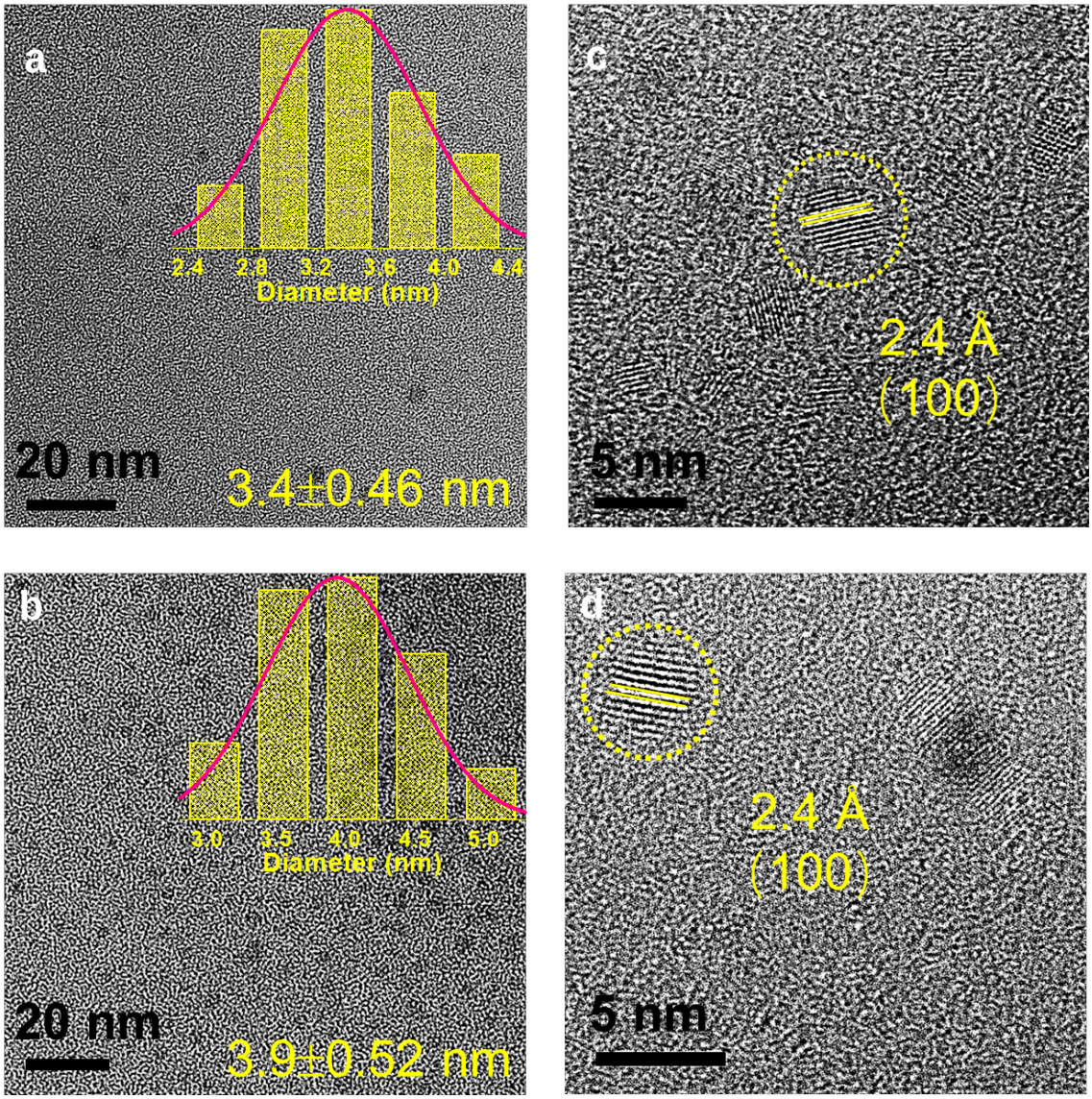


Figure S5 (a and c) TEM of CD-OH and CD-NH_2, (b-d) HR-TEM of CD-OH and CD-NH_2.

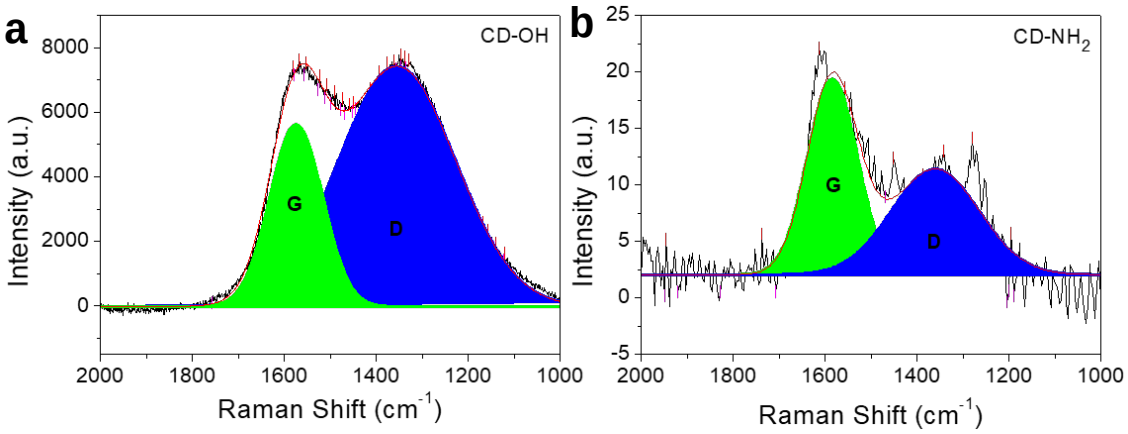


Figure S6 Raman spectra of CD-OH (a) and CD-NH_2 (b).

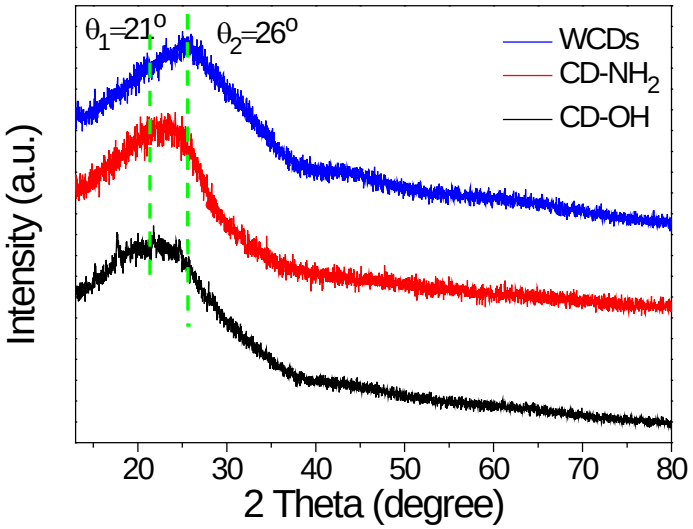


Figure S7 XRD pattern of CD-OH, CD-NH_2 and WCDs.

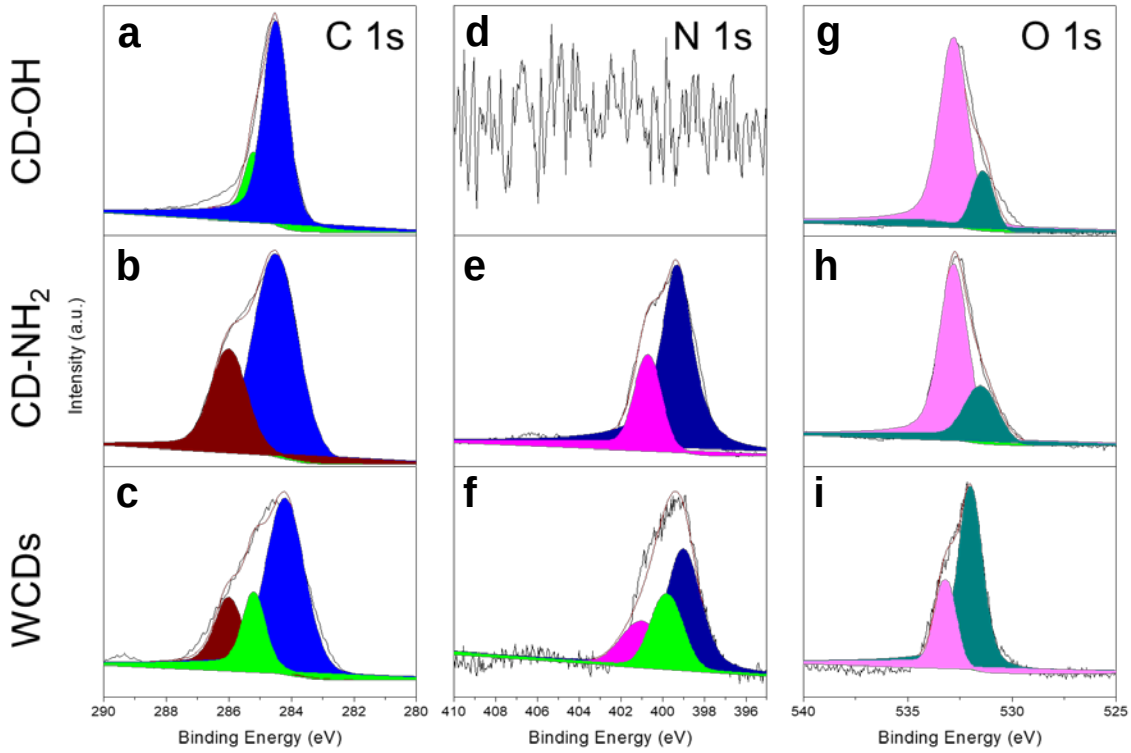


Figure S8 (a-c) The high-resolution C1s XPS spectra, (d-f) the high-resolution N1s XPS spectra, (g-i) the high-resolution O1s XPS spectra of CD-OH, CD-NH_2 and white light emitting CDs.

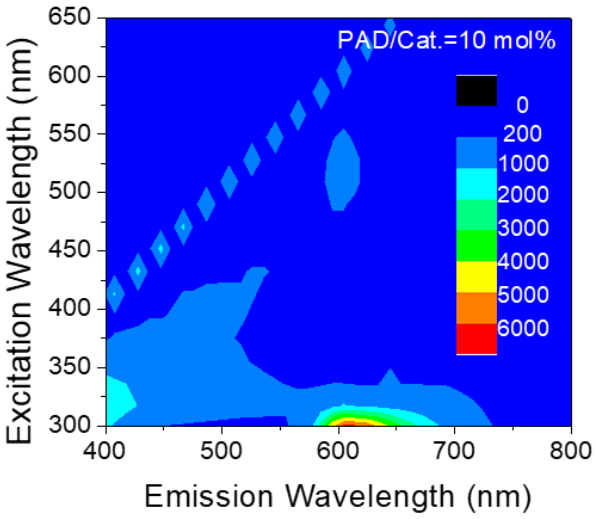


Figure S9 The 2D EEM spectra of CDs prepared from with different amount of the PDA in the catechol, the molar fraction of the PDA is 10 mol%.

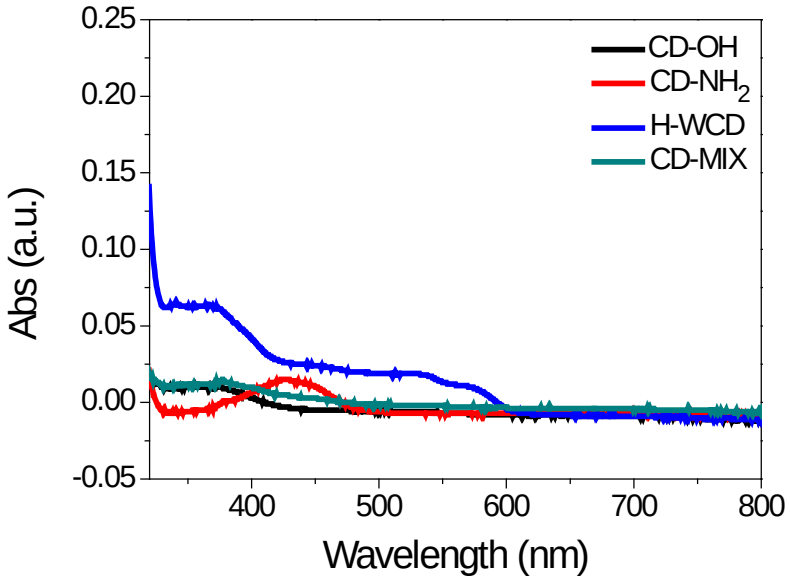


Figure S10 UV-vis spectra of CD-OH, CD-NH_2, white light emitting CD and the mixture of CD-OH and CD-NH_2.

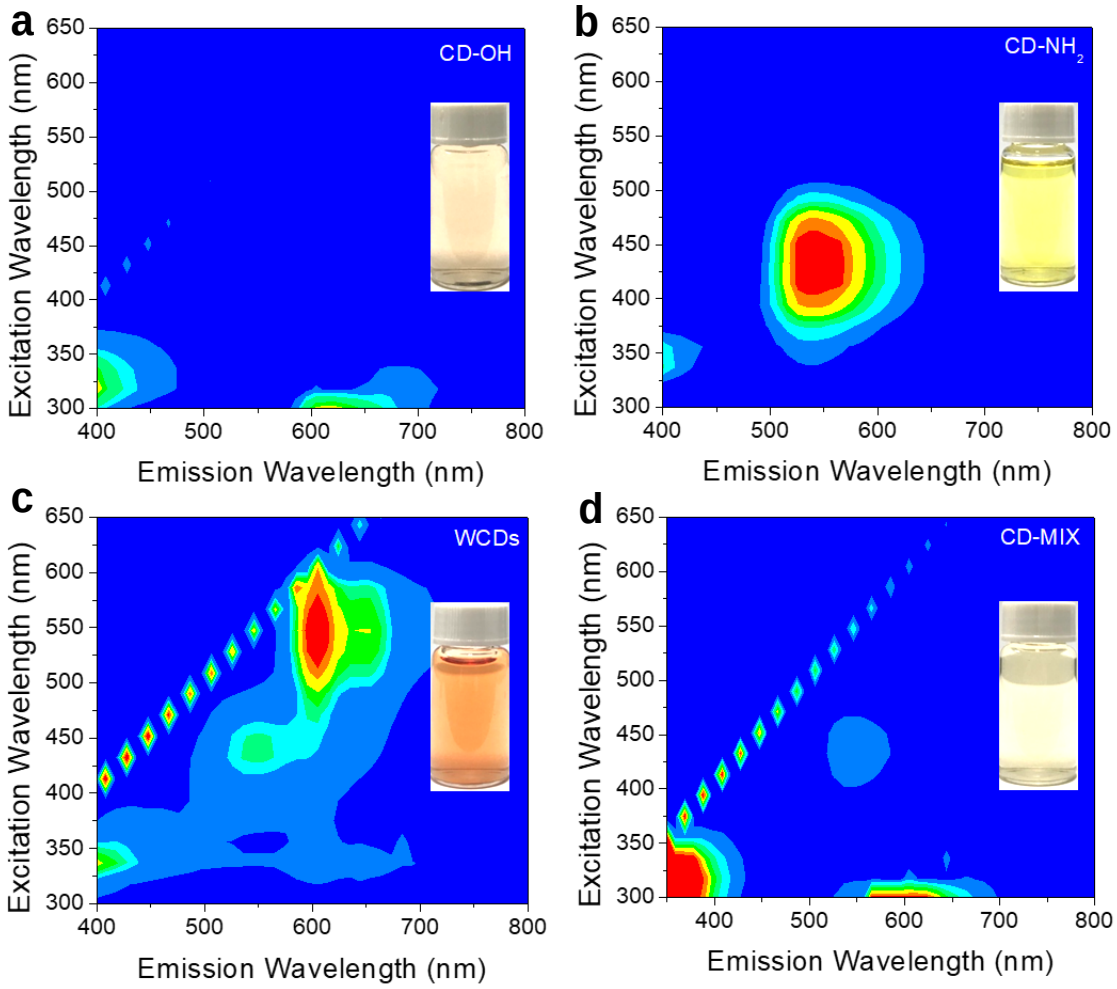


Figure S11 2D excitation-emission matrix of the CD-OH (a), CD-NH_2 (b), white light emitting CDs (c) and the mixture of CD-OH and CD-NH_2 (d).

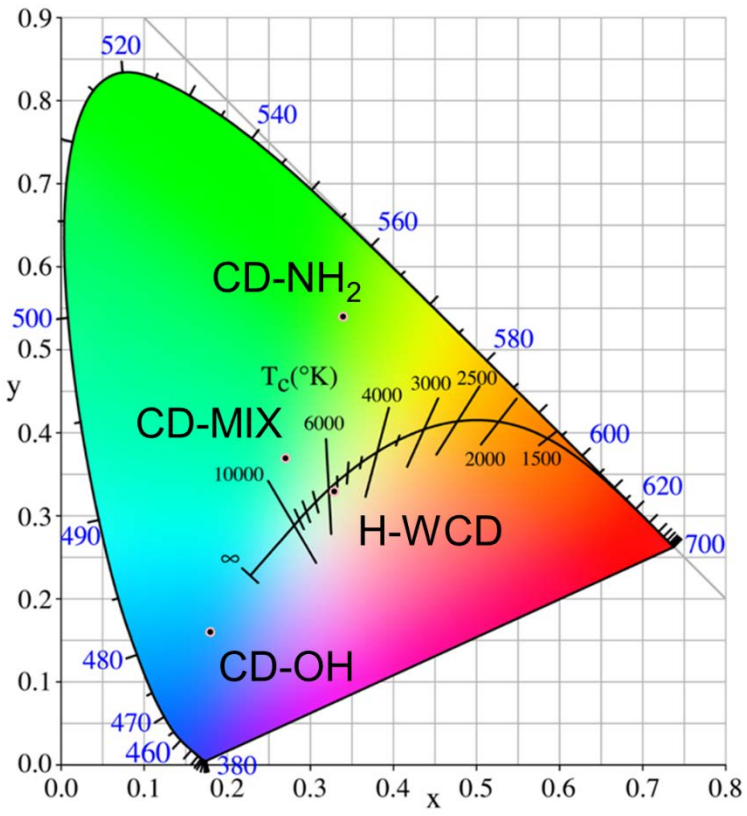


Figure S12 The CIE color coordinates of CD-OH, CD-NH_2, white light emitting CD and the mixture of CD-OH and CD-NH_2.

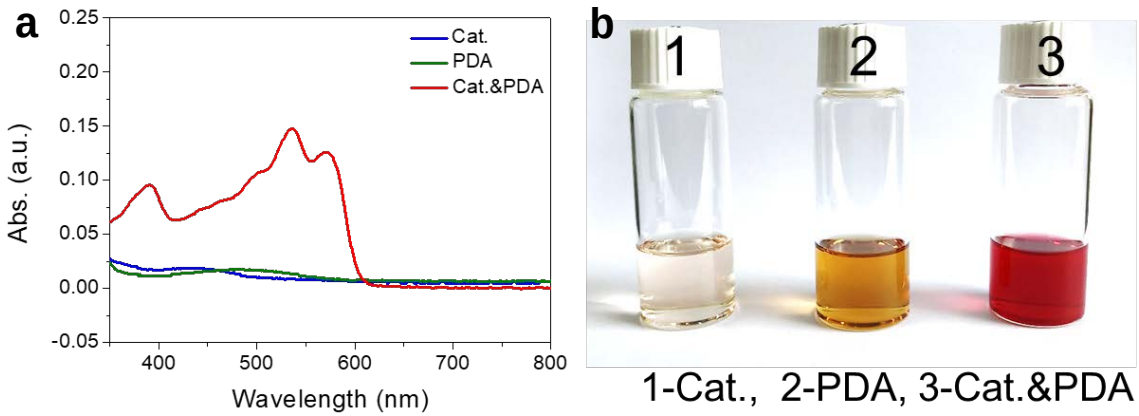


Figure S13 UV-vis spectrum (a) and photos (b) of the reactants dissolved in the NAA.

Table S4 CD-OH, CD-NH_2, white light emitting CD and the mixture of CD-OH and CD-NH_2.

	CD-OH	CD-NH_2	WCD	MIX
x	0.18	0.34	0.33	0.27
y	0.16	0.54	0.33	0.37
CCT [K]	558632	5436	5453	8307

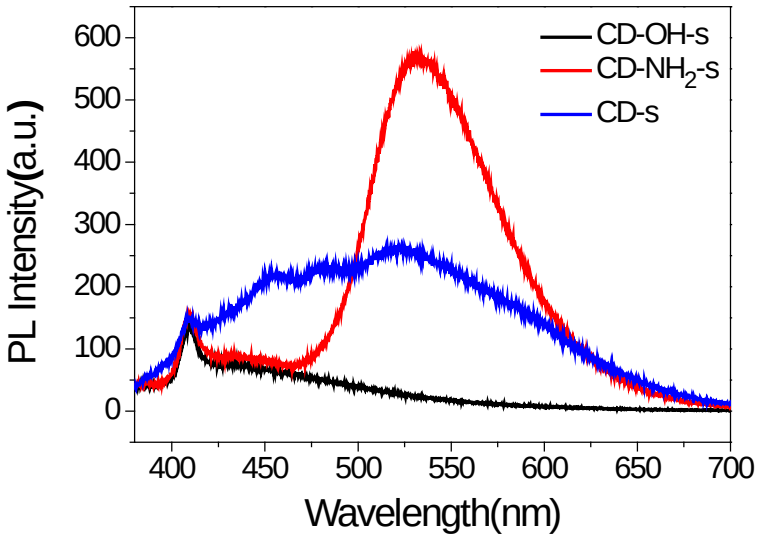


Figure S14 PL spectra under the excitation wavelength at 365 nm of the three kinds of CDs (CD-OH-s, CD-NH_2-s and CDs) with the slowly heating process.

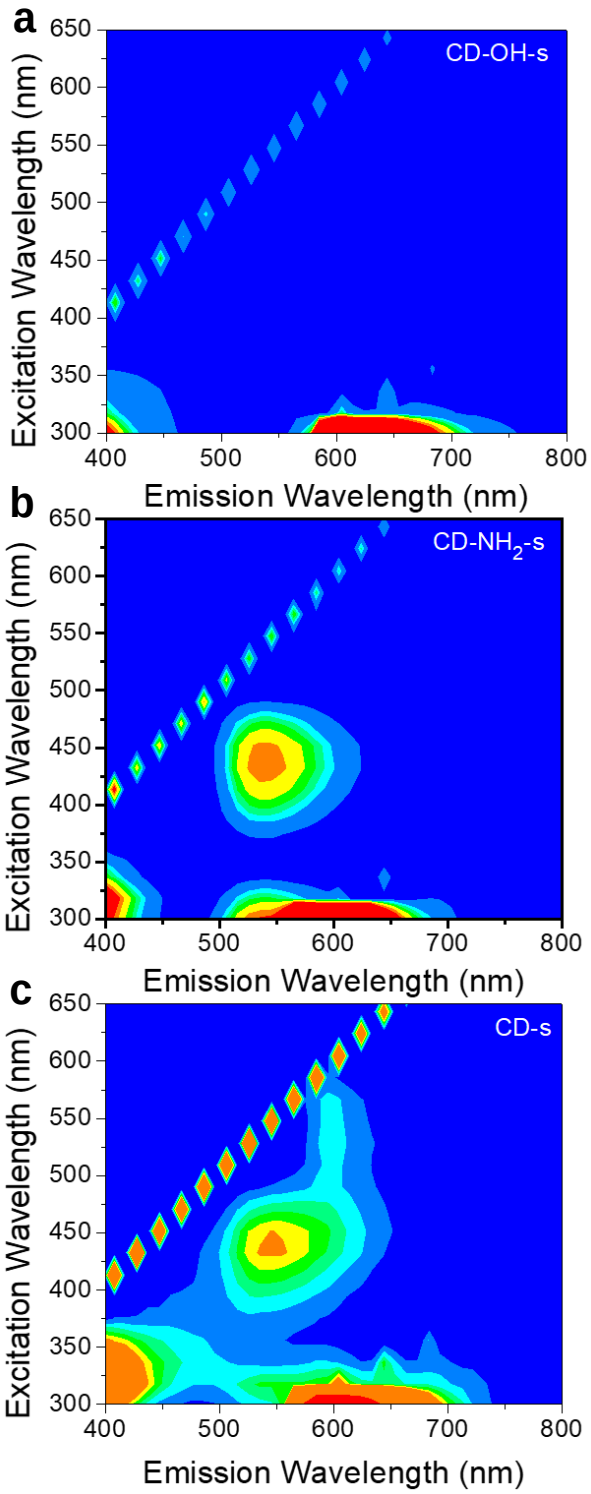


Figure S15 2D excitation-emission matrix of the CD-OH-s (a), CD-NH_2-s (b) and CDs (c).

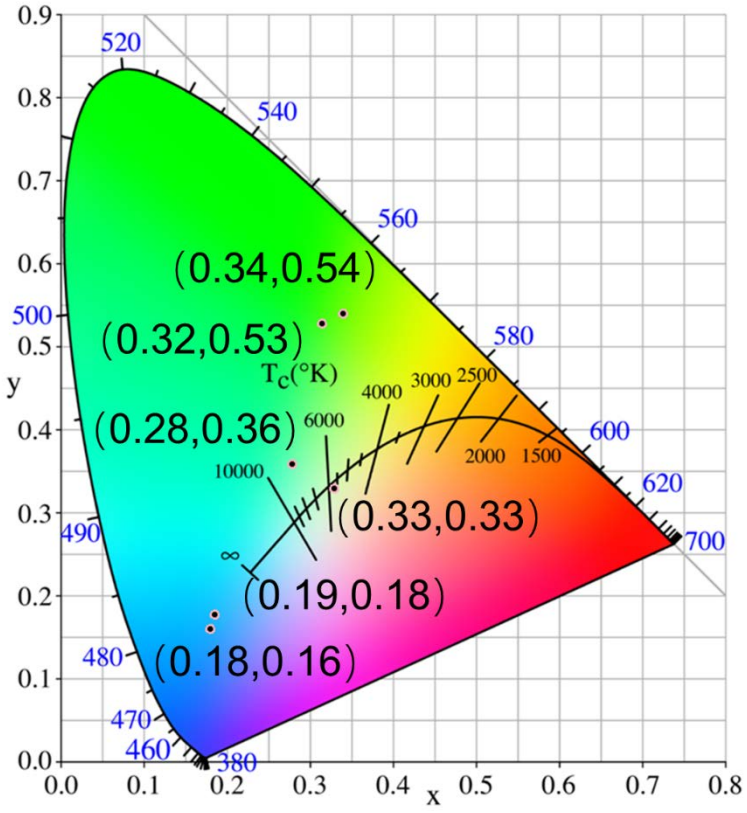


Figure S16 The CIE color coordinates of CD-OH (0.18,0.16), CD-OH-s (0.19,0.18), CD-NH_2 (0.34,0.54), CD-NH_2-s (0.32,0.53), CD-s (0.28,0.26) and white light emitting CDs (0.33,0.33).

Table S5 The CIE color coordinates and CCT of CD-OH, CD-OH-s, CD-NH_2, CD-NH_2-s, CD-s and white light emitting CDs.

	CD-OH	CD-OH-s	CD-NH_2	CD-NH_2-s	WCD	CD-s
x	0.18	0.19	0.34	0.32	0.33	0.28
y	0.16	0.18	0.54	0.53	0.33	0.36
CCT [K]	558632	99105504	5436	5839	5453	7940

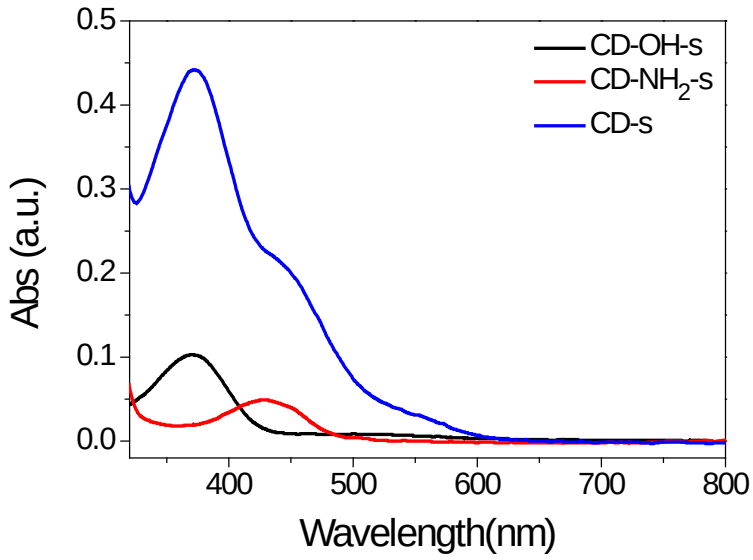
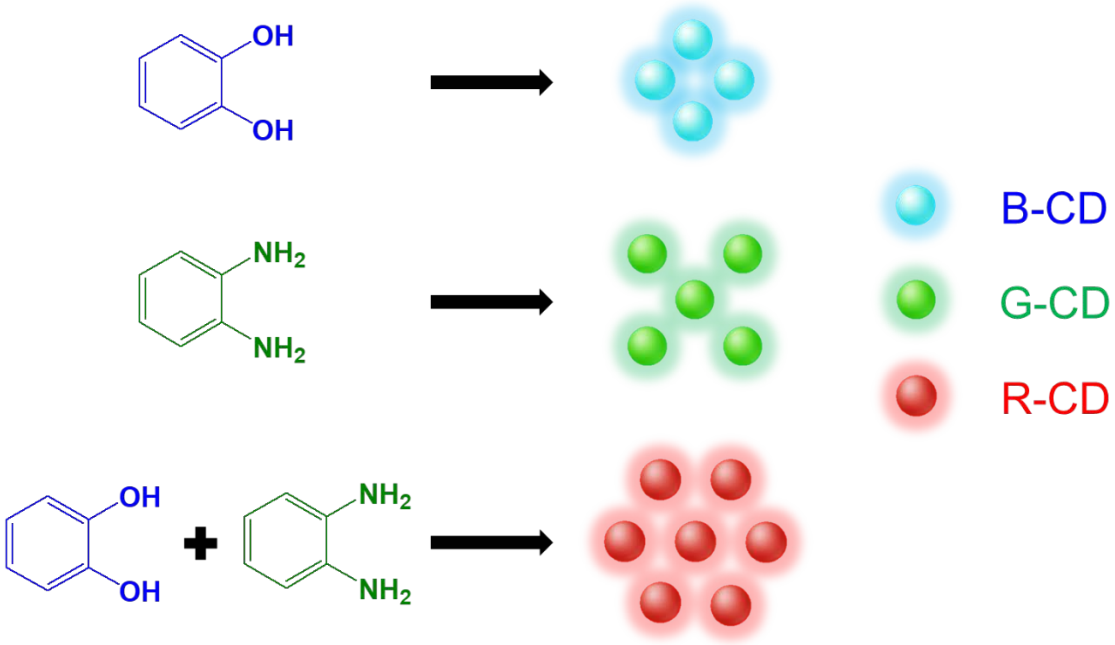


Figure S17 UV-vis spectra of the three kinds of CDs (CD-OH-s, CD-NH_2-s and CD-s) with the slowly heating process.



Scheme S1 The catechol self-carbonizes to form B-CDs, the G-CDs are prepared from the carbonization of the pure PDA. The R-CDs are synthesized from the complexes RP.

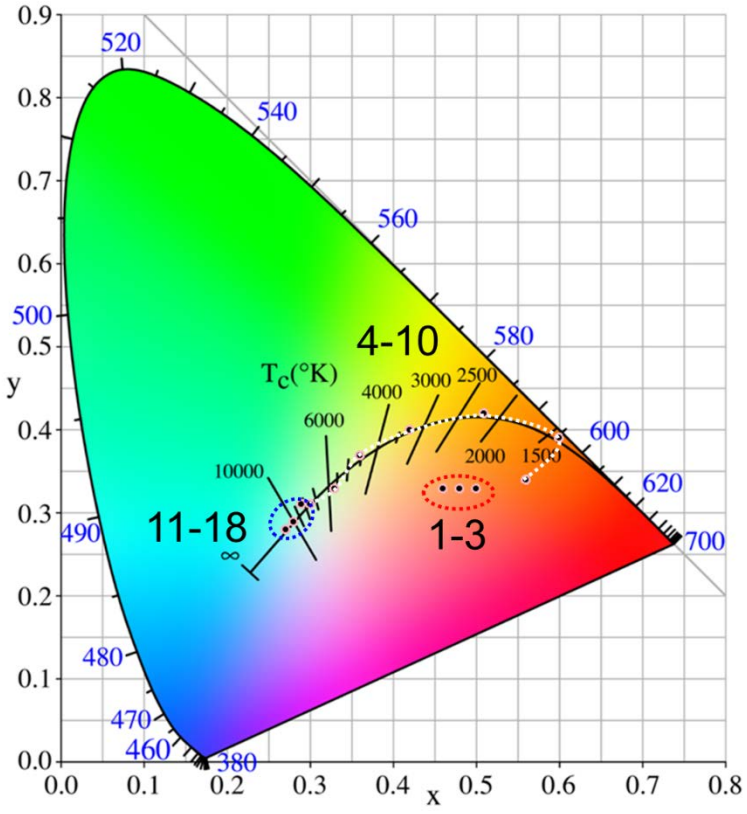
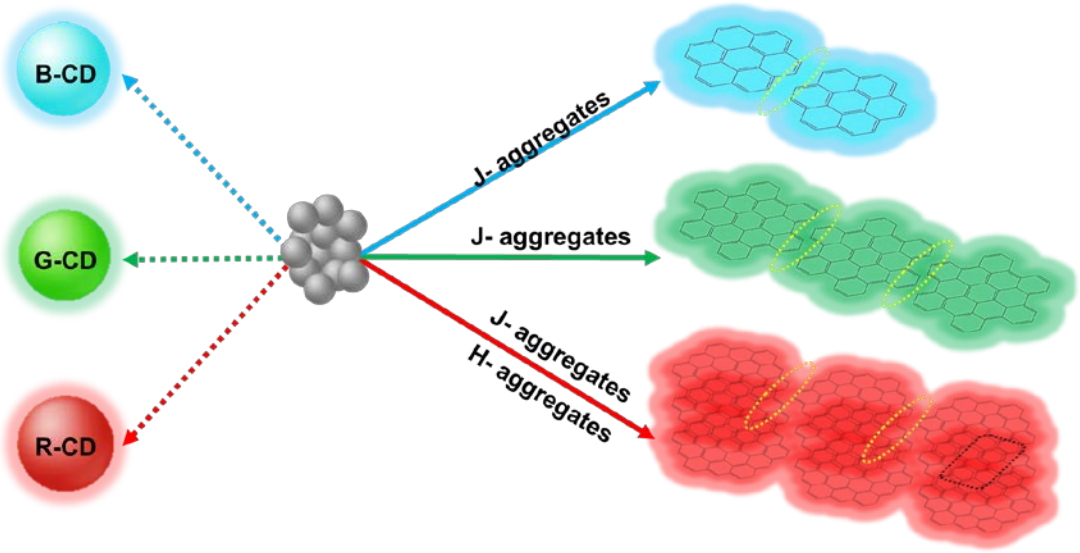


Figure S18 The CIE color coordinates of white light emitting CDs with different concentration (**Table S6**).

Table S6 The CIE color coordinates of white light emitting CDs with different concentration**.**

Line	C [mg ml^{-1}]
1	3.00
2	2.50
3	2.00
4	1.50
5	1.00
6	0.80
7	0.60
8	0.50
9	0.40
10	0.20
11	0.10
12	0.08
13	0.06
14	0.05
15	0.04
16	0.03
17	0.02
18	0.01



Scheme S2 The B-, G-CDs mainly form J-aggregates and R-CDs form both H- and J-aggregates in the high concentration.

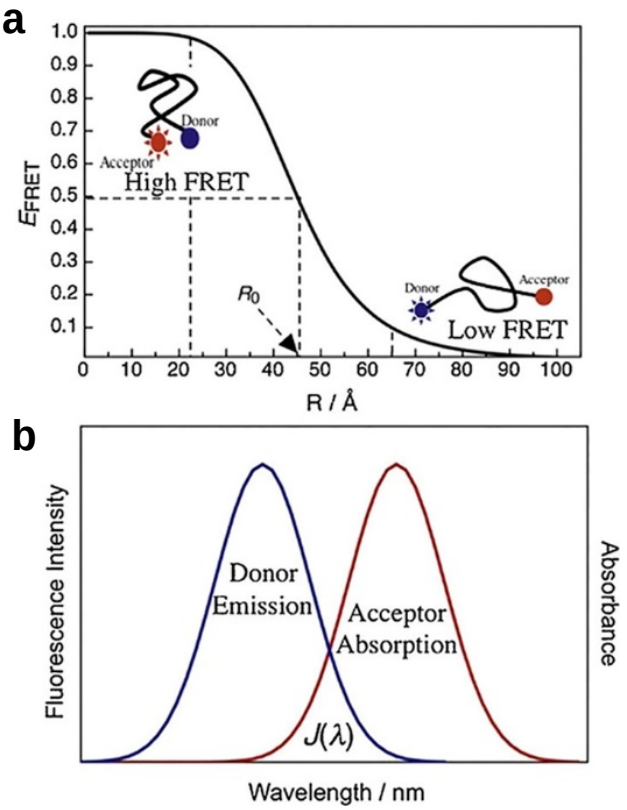


Figure S19 (a) Dependency of FRET efficiency (E_{FRET}) on the distance between FRET pair (R). The dashed area in the curve (E_{FRET} vs R) represents the sensitive FRET region for the FRET pair. (b) Graphical representation of spectral overlapping, J(λ), between donor fluorescence/emission spectra (blue) and acceptor absorption spectra (red). [42,43]

Table S7. The PL QY of CDs in ethanol at different concentration.

C [mg mL^{-1}]	B-CDs Ex:365 nm	G-CDs Ex:450 nm
3.5	32	50
3.0	35	52
2.5	37	53
2.0	31	49
1.5	20	45
1.0	14	41

Table S8. The PL QY of the composites (B-, G-CDs/silica resin).

C [mg mL^{-1}]	B-CDs Ex:365 nm	G-CDs Ex:450 nm
1.0	40	74
0.75	43	75
0.5	42	73
0.25	41	73
0.05	40	72

Table S9 The corresponding CIE 1931 and their average PL decay lifetime of CD-OH, CD-NH_2 and white light emitting CDs at 0.5 mg mL^{-1}, and white light emitting CDs at and 0.05 mg mL^{-1}, respectively.

	CD-OH-0.5	CD-NH_2-0.5	WCD-0.05	WCD-0.5
CIE	(0.18,0.16)	(0.34,0.54)	(0.27,0.29)	(0.33,0.33)
450	4.23	0	4.22	2.32
520	0	6.10	5.96	11.89
600	0	0	3.84	13.51

Table S10 The CIE color coordinates of white light emitting CDs with different concentration**.**

No.	C [mg ml^{-1}]
1	0.30
2	1.00
3	1.50
4	2.00
5	2.25
6	2.50
7	2.75
8	3.00
9	3.50
10	4.00
11	4.50
12	5.00

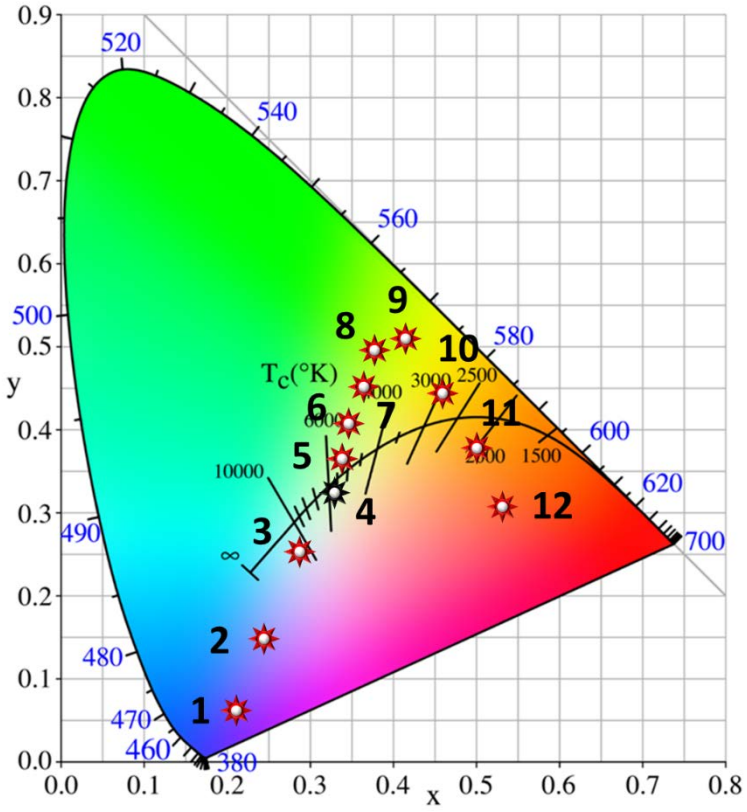


Figure S20 The corresponding CIE 1931 coordinates for these 12 devices.

Figure S21 The photograph images of the white plane light source in off state.

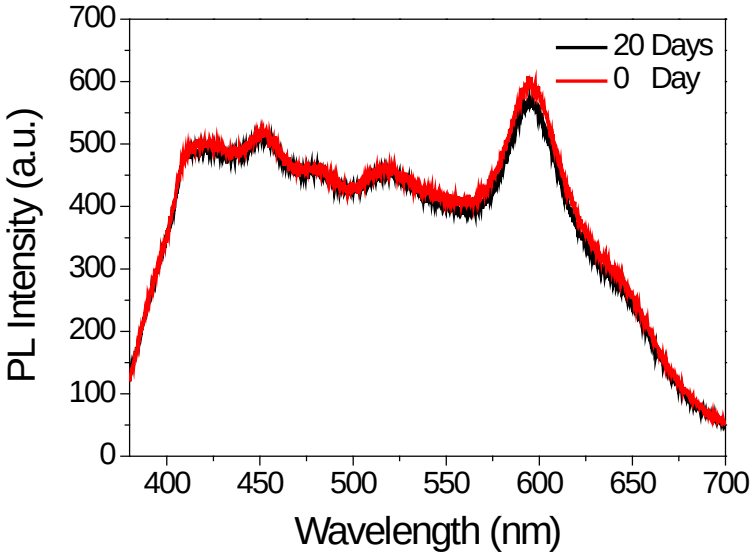


Figure S22 The emitting spectra of the white LED at initial and after illustrating over 20 days.