One-Step Microwave Synthesis of N-Doped Hydroxyl-Functionalized Carbon Dots with Ultra-High Fluorescence Quantum Yields

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The QYs were determined by slope method using the reference of quinine sulfate (0.1 M, H_2SO_4 as solvent, excitation wavelength at 350 nm, QY=56%): CDs were dissolved in ultrapure water to form several kinds of different concentrations dilute solutions, the absorbance and the fluorescence spectra (350 nm excitation) of the same solution were measured, then the integrated photoluminescence intensity and the absorbance value (less than 0.1 at 350 nm wavelength) were plotted to give the curve of the CDs samples with that of the references. The absolute QY values were calculated using the following equation:

$$\Phi_x=\Phi_s(K_x/K_s)(\eta_x/\eta_s)^2 \quad (1)$$

where Φ is the QY, K is the slope determined by the curves and η is the refractive index. The subscript "s" refers to quinine sulfate and "x" refers to the CDs. For these aqueous solutions, $\eta_x/\eta_s=1$. So the equation was simplified to:

$$\Phi_x=\Phi_s(K_x/K_s) \quad (2)$$

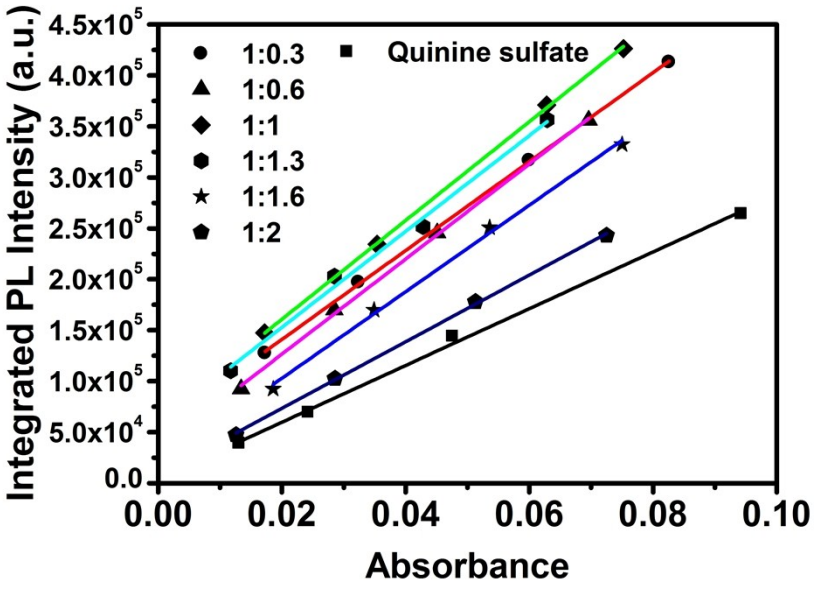

Figure S1. The QYs of Tris-CDs obtained by the slope method.

Table S1. The QYs values of Tris-CDs.

Sample	1:X (mole ratio)	K (slope)	Φ (QY)
Quinine sulfate	N/A	2.68E6	56 %
Tris-CD(1:0.3)	0.3	4.36E6	91.1 %
Tris-CD(1:0.6)	0.6	4.57E6	95.5 %
Tris-CD(1:1)	1.0	4.75E6	99.3 %
Tris-CD(1:1.3)	1.3	4.61E6	96.3 %
Tris-CD(1:1.6)	1.6	4.29E6	89.6 %
Tris-CD(1:2)	2.0	3.27E6	68.3 %

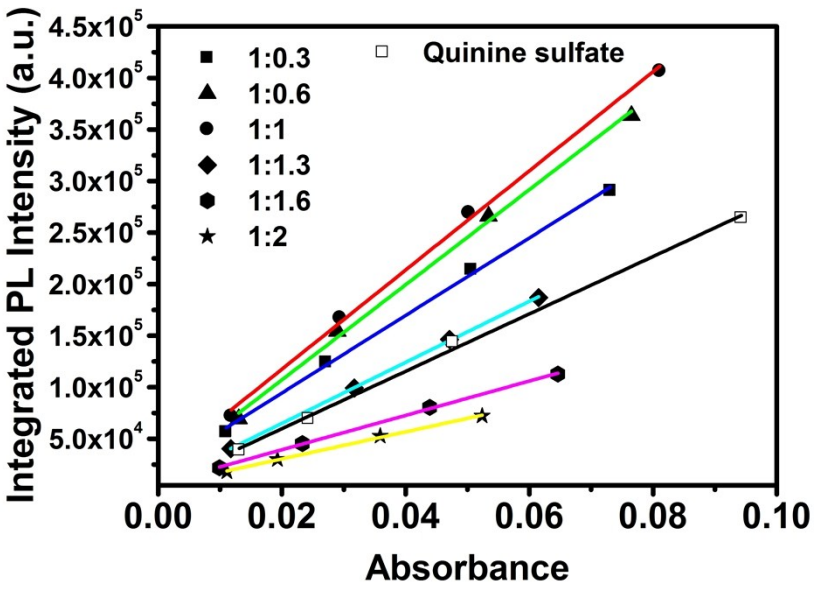

Figure S2. The QYs of EA-CDs obtained by the slope method.

Table S2. The QYs values of EA-CDs.

Sample	1:X (mole ratio)	K (slope)	Φ (QY)
Quinine sulfate	N/A	2.68E6	56 %
EA-CD(1:0.3)	0.3	3.76E6	78.6 %
EA-CD(1:0.6)	0.6	4.42E6	92.4 %
EA-CD(1:1)	1.0	4.61E6	96.3 %
EA-CD(1:1.3)	1.3	2.96E6	61.9 %
EA-CD(1:1.6)	1.6	1.67E6	34.9 %
EA-CD(1:2)	2.0	1.31E6	27.4 %

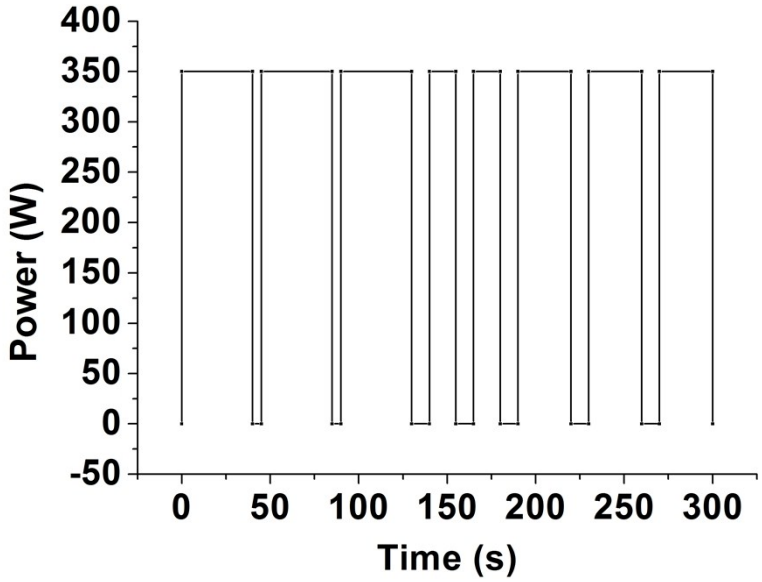

Figure S3. The heating curve of power against time in microwave synthesis.

Table S3. The yields of Tris-CDs and EA-CDs.

Sample	Molar ratio (1:X)					
	1:0.3	1:0.6	1:1	1:1.3	1:1.6	1:2
Tris-CDs	0.858	0.871	0.881	0.872	0.863	0.815
EA-CDs	0.793	0.799	0.807	0.725	0.698	0.640

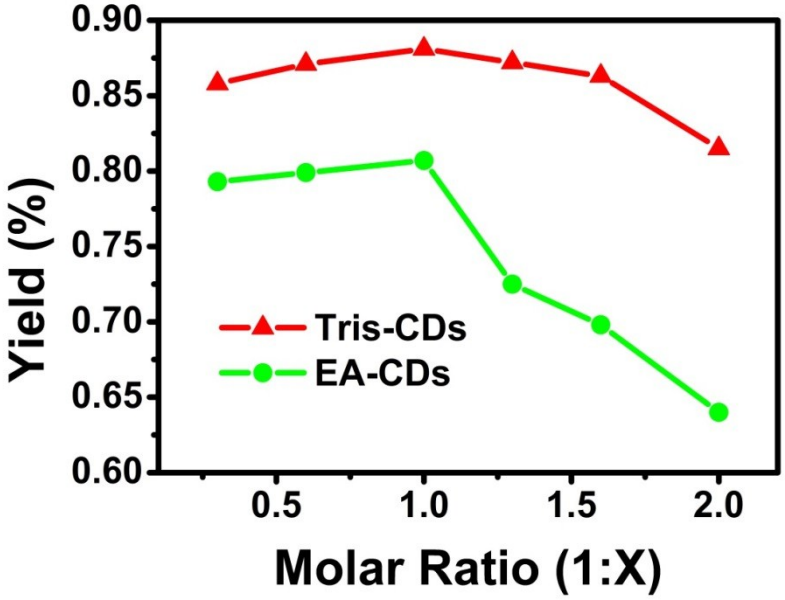

Figure S4. The yields of Tris-CDs and EA-CDs.

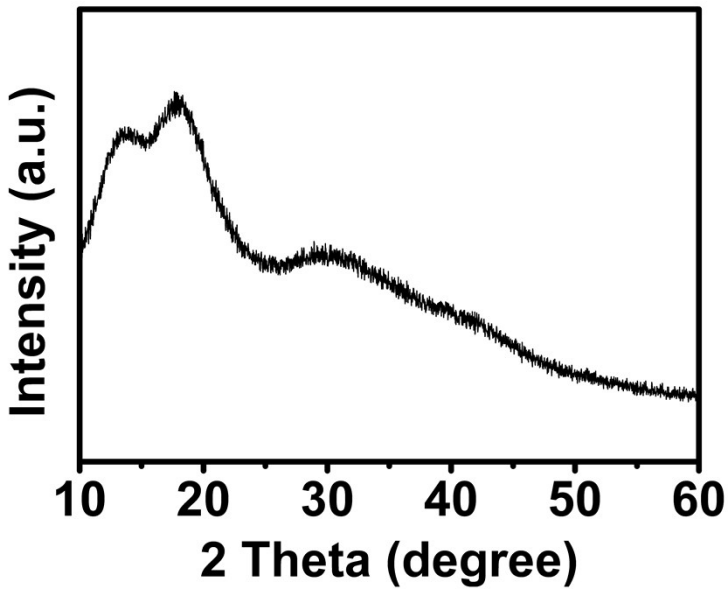

Figure S5. The XRD spectrum of Tris-CD(1:1).

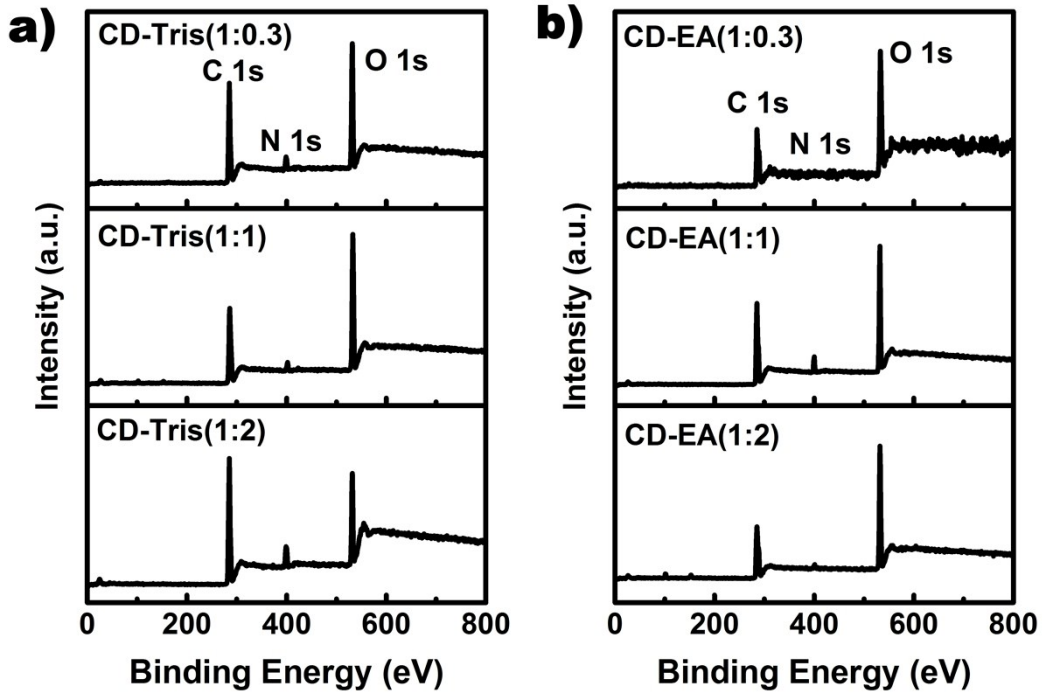

Figure S6. a) The full scan XPS spectra of Tris-CD(1:0.3) (above), Tris-CD(1:1) (middle), and Tris-CD(1:2) (below), b) The full scan XPS spectra of EA-CD(1:0.3) (above), EA-CD(1:1) (middle), and EA-CD(1:2) (below).

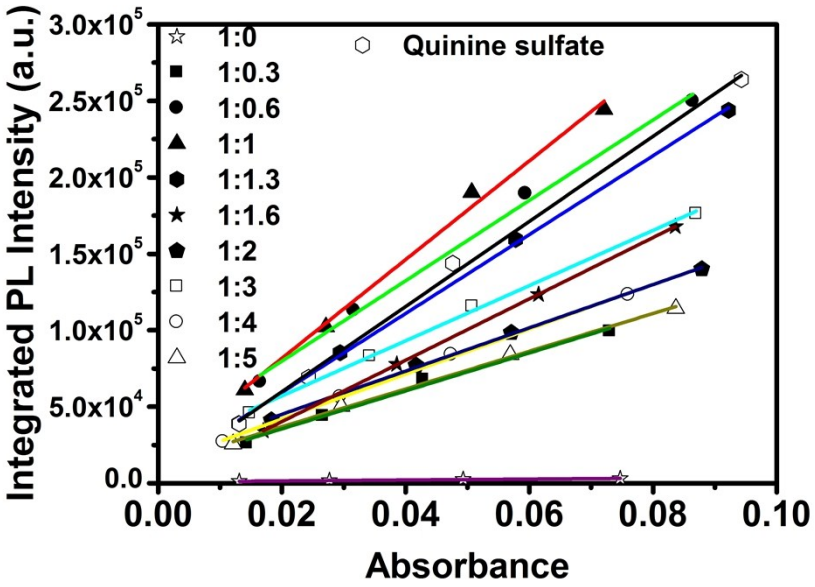

Figure S7. The QYs of ED-CDs obtained by the slope method.

Table S4. The QYs values of ED-CDs.

Sample	1:X (mole ratio)	K (slope)	Φ (QY)
Quinine sulfate	N/A	2.68E6	56 %
ED-CD(1:0)	0	30462.14	0.6 %
ED-CD(1:0.3)	0.3	1.24E6	25.9 %
ED-CD(1:0.6)	0.6	2.63E6	55.0 %
ED-CD(1:1)	1.0	3.23E6	67.5 %
ED-CD(1:1.3)	1.3	2.58E6	53.9 %
ED-CD(1:1.6)	1.6	2.01E6	42.0 %
ED-CD(1:2)	2.0	1.41E6	29.5 %
ED-CD(1:3)	3.0	1.80E6	37.6 %
ED-CD(1:4)	4.0	1.47E6	30.7 %
ED-CD(1:5)	5.0	1.23E6	25.7 %

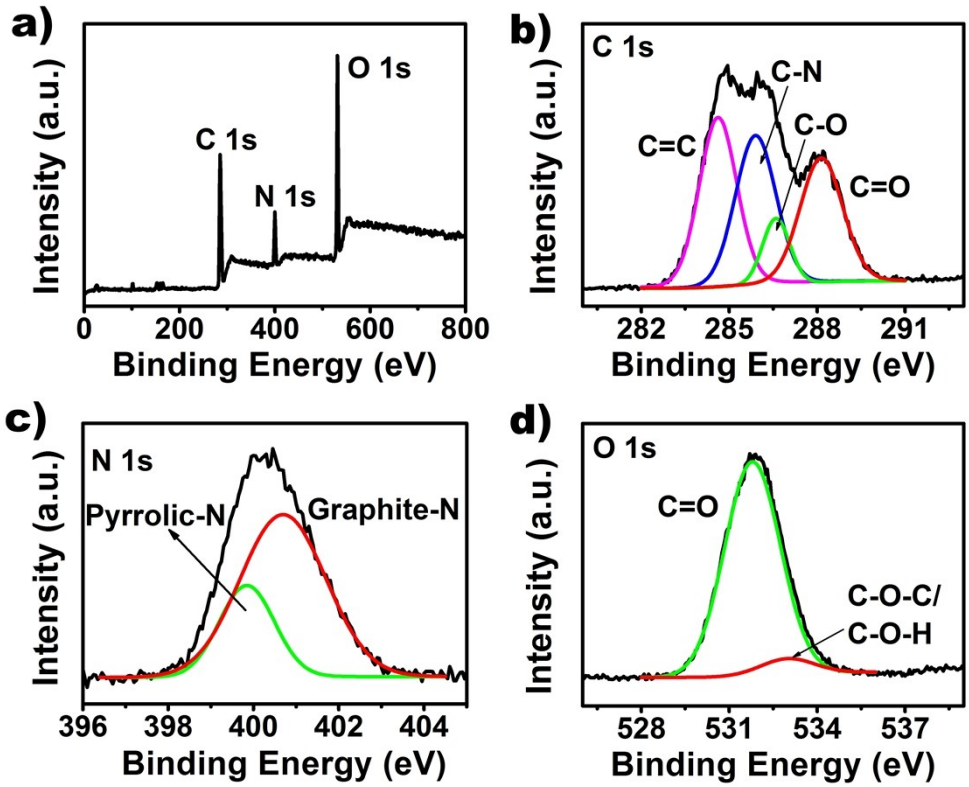

Figure S8. a) The full XPS spectrum of ED-CD(1:1), b) C 1s, c) N1s, and d) O1s spectra of ED-CD(1:1).

Table S5. The contents of various chemical bonds of ED-CD(1:1) in XPS spectra.

C 1s				N 1s	
C=C	C-N	C-O	C=O	pyrrolic-N	graphite-N
0.33	0.31	0.09	0.27	0.05	0.13

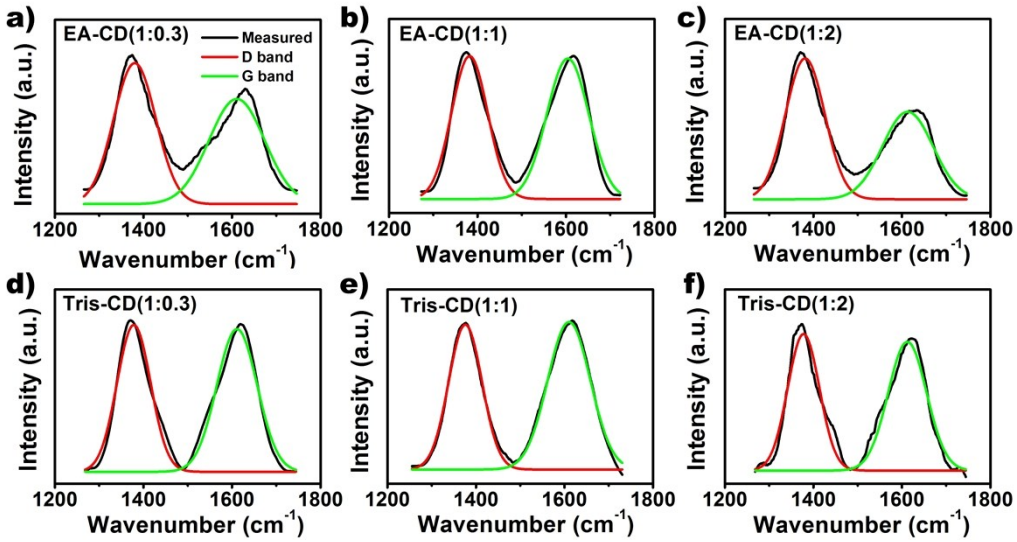

Figure S9. The Raman spectra of a) EA-CD(1:0.3), b) EA-CD(1:1), EA-CD(1:2), d) Tris-CD(1:0.3), e) Tris-CD(1:1) and f) Tris-CD(1:2).

Table S6. The values of I_D/I_G of ED-CDs and Tris-CDs.

Tris-CD (1:0.3)	Tris-CD (1:1)	Tris-CD (1:2)	EA-CD (1:0.3)	EA-CD (1:1)	EA-CD (1:2)
1.03	0.98	1.06	1.38	1.01	1.62

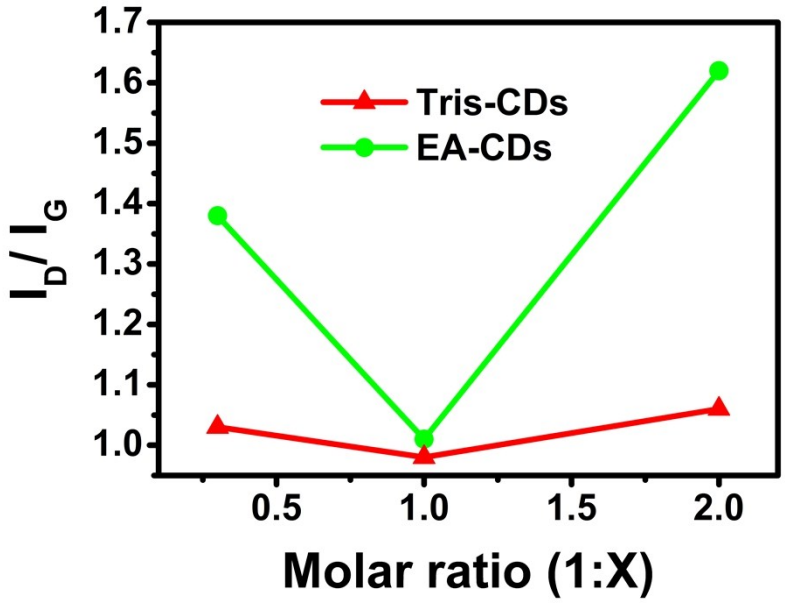

Figure S10. The dependence of the values of I_D/I_G of ED-CDs and Tris-CDs on molar ratio.

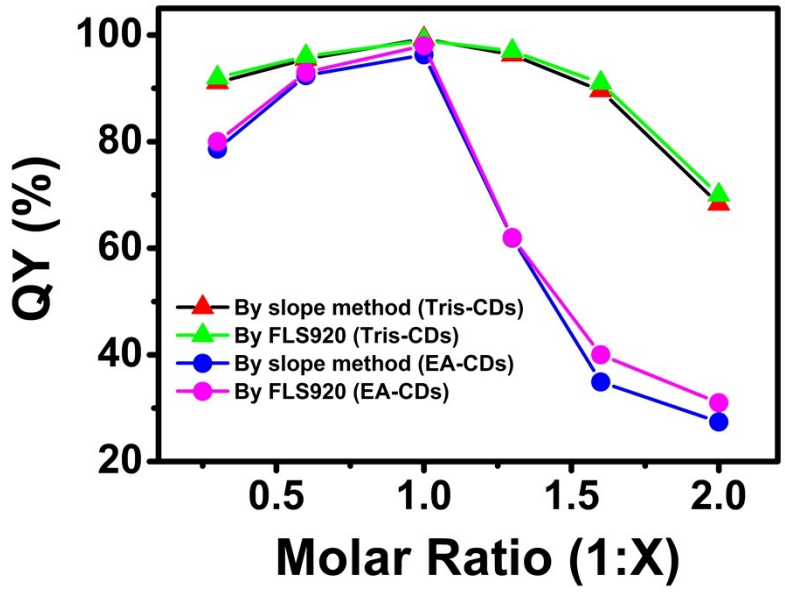

Figure S11. Comparison of relative and absolute fluorescent QYs, measured by slope method and FLS920 fluorescence spectrophotometer, respectively.

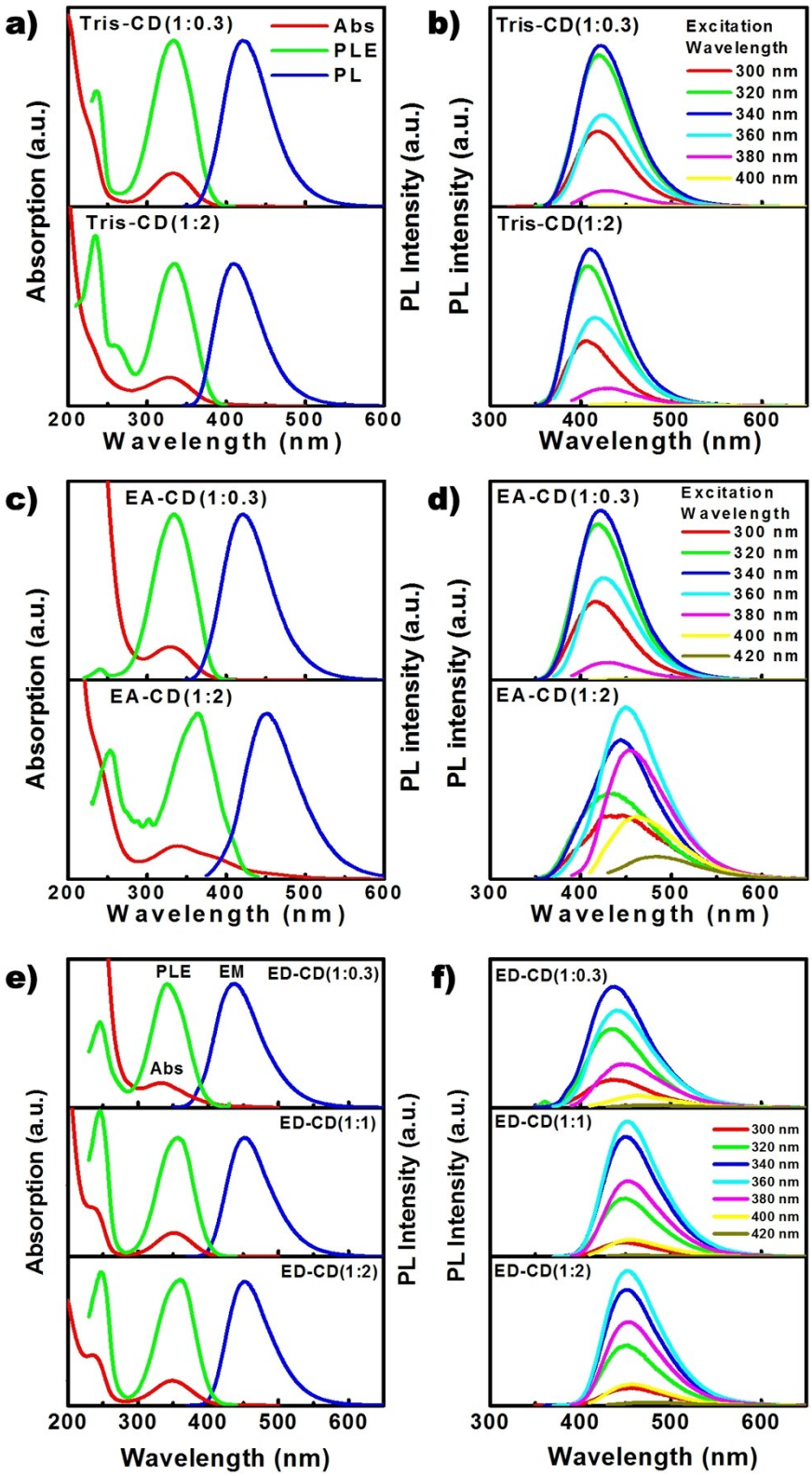

Figure S12. UV/Vis absorption (red line), PL excitation (green line), and emission (blue line) spectra of a) Tris-CD(1:0.3) and Tris-CD(1:2), c) EA-CD(1:0.3) and EA-CD(1:2), e) ED-CDs in aqueous solutions. PL emission spectra of b) Tris-CD(1:0.3) and Tris-CD(1:2), d) EA-CD(1:0.3) and EA-

CD(1:2), f) ED-CD aqueous solutions at different excitation wavelengths of 300 nm (red), 320 nm (green), 340 nm (blue), 360 nm (cyan), 380 nm (magenta), 400 nm (yellow), 420 nm (deep yellow).

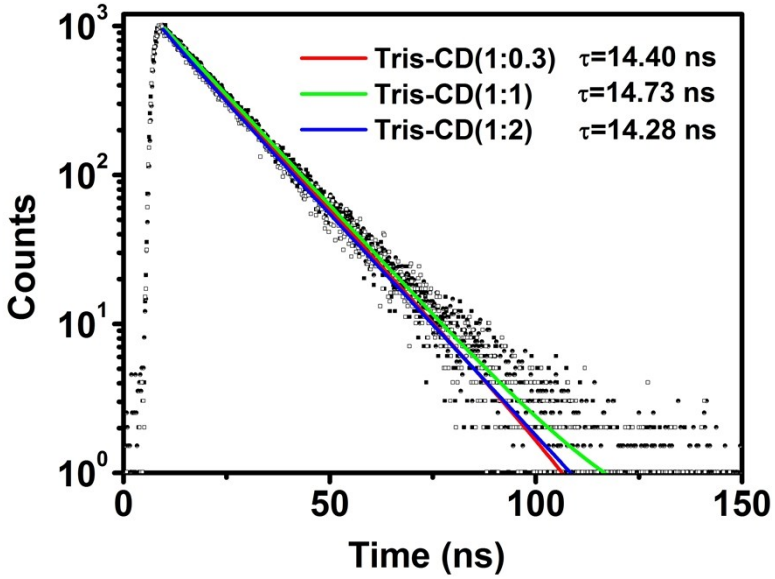

Figure S13. PL decay curves of Tris-CD(1:0.3) (red line), Tris-CD(1:1) (green line) and Tris-CD(1:2) (blue line).

Table S7. The lifetime (τ), decay fitting, QY and PL behavior of Tris-CD(1:0.3), Tris-CD(1:1) and Tris-CD(1:2).

Sample	Tris-CD(1:0.3)	Tris-CD(1:1)	Tris-CD(1:2)
τ_1 (ns) /proportion	14.40/100%	12.68 /36.8%	7.03 /4.8%
τ_2 (ns) /proportion	N/A	15.92 /63.2%)	14.65/95.2%
χ^2	0.976	0.997	1.079
τ_{ave} (ns)	14.40	14.73	14.28
QY	91.1%	99.3%	68.3%

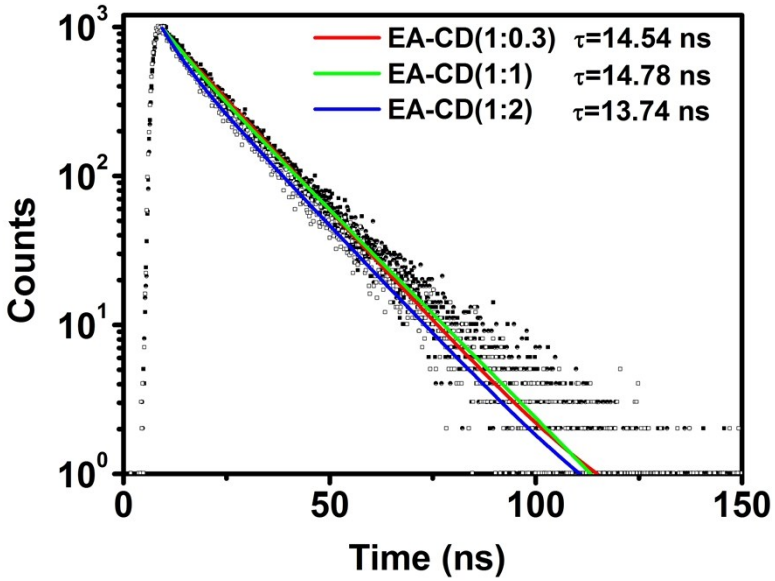

Figure S14. PL decay curves of EA-CD(1:0.3) (red line), EA-CD(1:1) (green line) and EA-CD(1:2) (blue line).

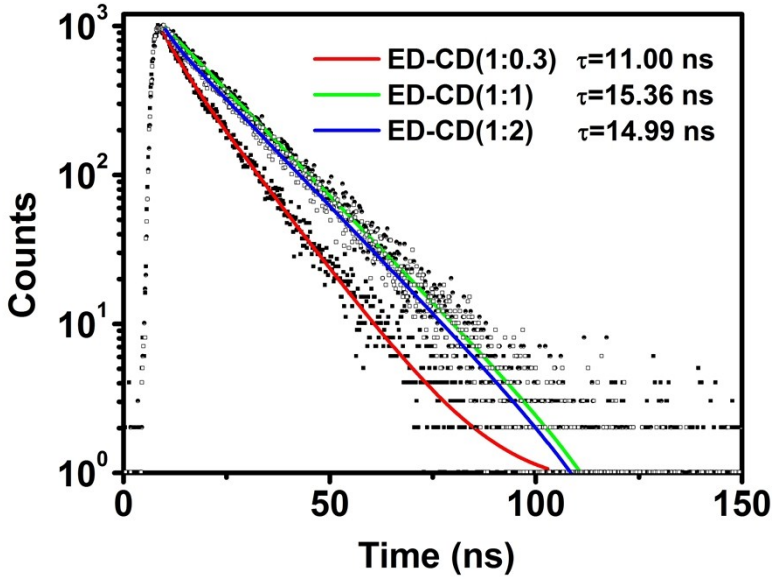

Figure S15. PL decay curves of ED-CD(1:0.3) (red line), ED-CD(1:1) (green line) and ED-CD(1:2) (blue line).

Table S8. The lifetime (τ), decay fitting, QY and PL behaviors of EA-CD(1:0.3), EA-CD(1:1) and EA-CD(1:2).

Sample	EA-CD(1:0.3)	EA-CD(1:1)	EA-CD(1:2)
τ_1 (ns) /proportion	14.54 ns (100%)	6.74 ns (9.35%)	4.83 ns (11.31%)
τ_2 (ns) /proportion	N/A	15.61 ns (90.65%)	14.88 ns (88.69%)
χ^2	1.013	1.003	1.003
τ (ns)	14.54	14.78	13.74
QY	78.6%	96.3%	27.4%

Table S9. The lifetime (τ), decay fitting, QY and PL behaviors of ED-CD(1:0.3), ED-CD(1:1) and ED-CD(1:2).

Sample	ED-CD(1:0.3)	ED-CD(1:1)	ED-CD(1:2)
τ_1 (ns) (proportion)	4.64 (12.82%)	15.36 (100%)	14.99 (100%)
τ_2 (ns) (proportion)	11.92 (87.18%)	N/A	N/A
χ^2	1.022	1.014	1.081
τ (ns)	11.00	15.36	14.99
QY	25.9%	67.5%	29.5%

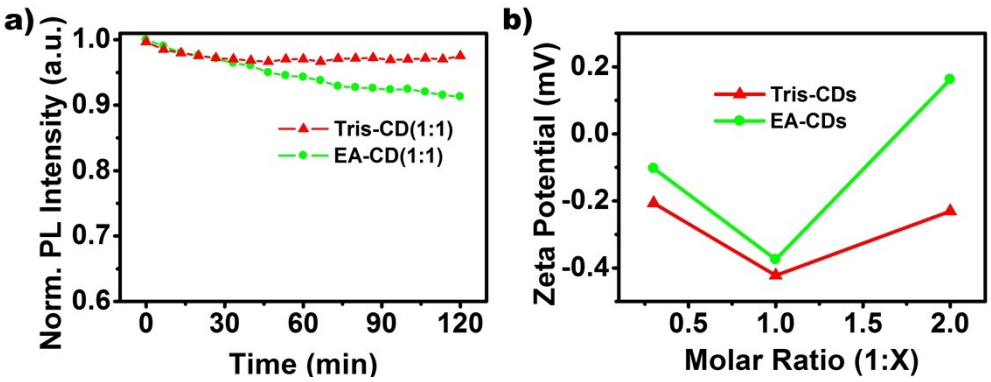

Figure S16. Resistance to photobleaching and zeta potential of Tris-CDs and EA-CDs.

Table S10. τ, QY, radiative transition rate (K_R) and non-radiative transition rate (K_{NR}) of ED-CDs.

Sample	Molar ratio (1:X)		
	1:0.3	1:1	1:2
τ	11.00 ns	15.36 ns	14.99 ns
QY	25.9%	67.5%	29.5%
K_R (s^{-1})	2.35×10^7	4.39×10^7	1.97×10^7
K_{NR} (s^{-1})	6.74×10^7	2.12×10^7	4.70×10^7
K_R/K_{NR}	0.35	2.07	0.42

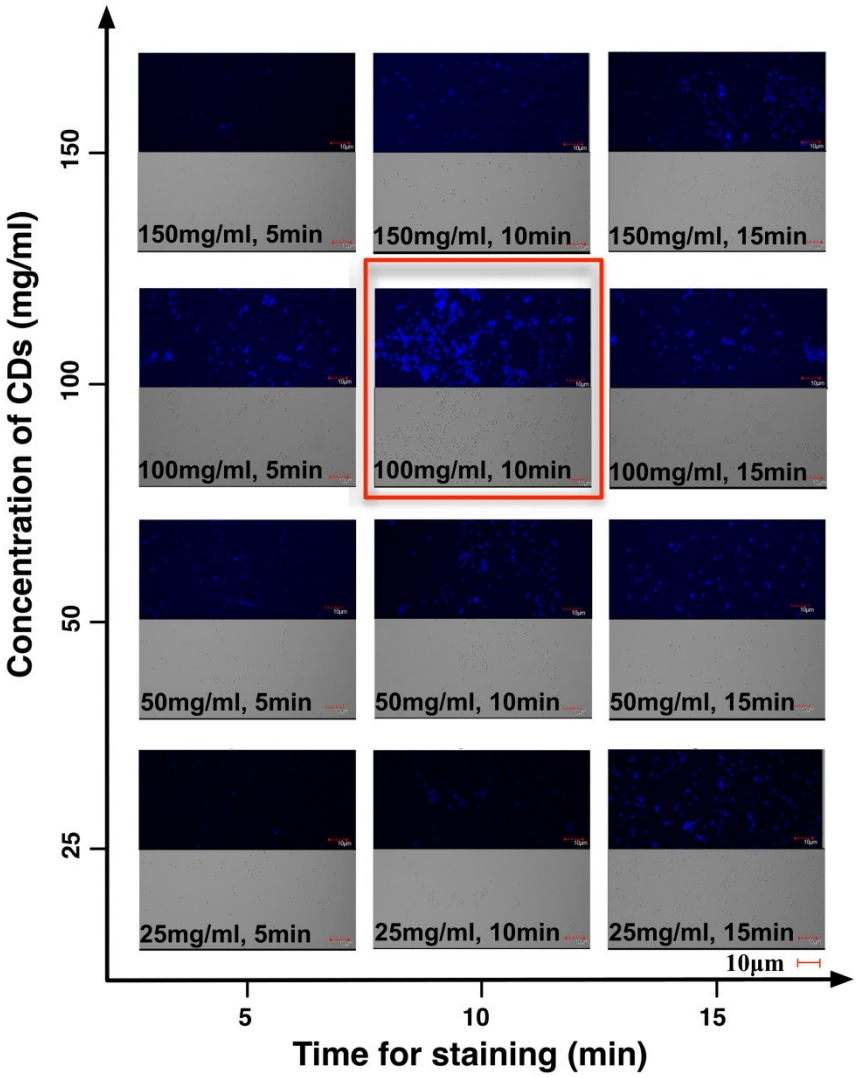

Figure S17. Optimization of the concentration of Tris-CD (1:1) and staining time. *E. coli* was used to optimize the concentration of Tris-CD (1:1) and the time for staining. CDs saline solution with the concentration of 25 mg/ml, 50 mg/ml, 100 mg/ml and 150 mg/ml were mixed with the suspensions of 10^8 cfu/ml *E. coli* pure culture (1:1) and incubated for 5 min, 10 min and 15 min, respectively. Then the stained bacteria were collected by centrifuging 6000 rpm (2391 (*g)) for 5 min and washed twice with saline. Finally, the staining effect was examined by the confocal microscopy.