

## Supplementary Information

### Preparation of multicolor-emissive carbon dots with high quantum yields and their epoxy composites for fluorescence anti-counterfeiting and light-emitting devices

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### Experimental

#### Quantum yield calculation

The relative quantum yields (QYs) of the CDs were calculated using Eq. (1),<sup>1-3</sup>

$$\varphi_x = \varphi_{std} \times \frac{I_x}{I_{std}} \times \frac{A_{std}}{A_x} \times \frac{\eta_x^2}{\eta_{std}^2} \quad (1)$$

where  $\varphi_x$  is the QY of the obtained CDs and  $\varphi_{std}$  is the quantum yield of the referenced fluorescent dyes.  $I_x$  and  $I_{std}$  are the integrated emission intensities of CDs and the referenced fluorescence dyes obtained using a fluorescent spectrometer.  $A$  is the absorbance of CDs and the referenced fluorescent dyes measured using a UV-vis spectrometer.  $\eta_x$  and  $\eta_{std}$  are the refractive indices (1.36 for ethanol) of the solvent.

Specifically, quinine sulfate (QY=55% in 0.1 M H<sub>2</sub>SO<sub>4</sub>) was selected as the reference in the emission range of 400-480 nm for B-CDs, rhodamine 6G (QY=95%

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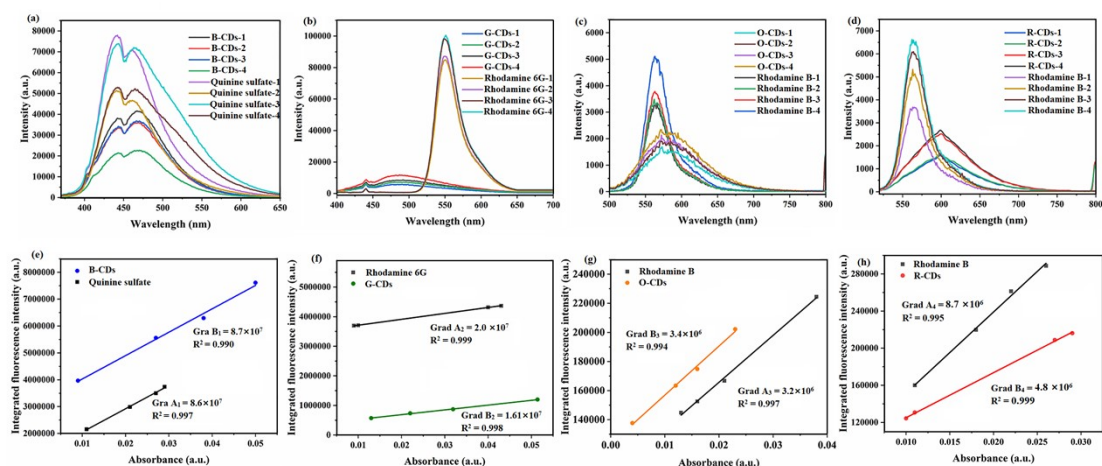
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in ethanol) as reference in the emission range of 480-560 nm for G-CDs, and rhodamine B (QY=56% in ethanol) as reference in the emission range of 580-610 nm for O-CDs and R-CDs.<sup>4,6</sup>

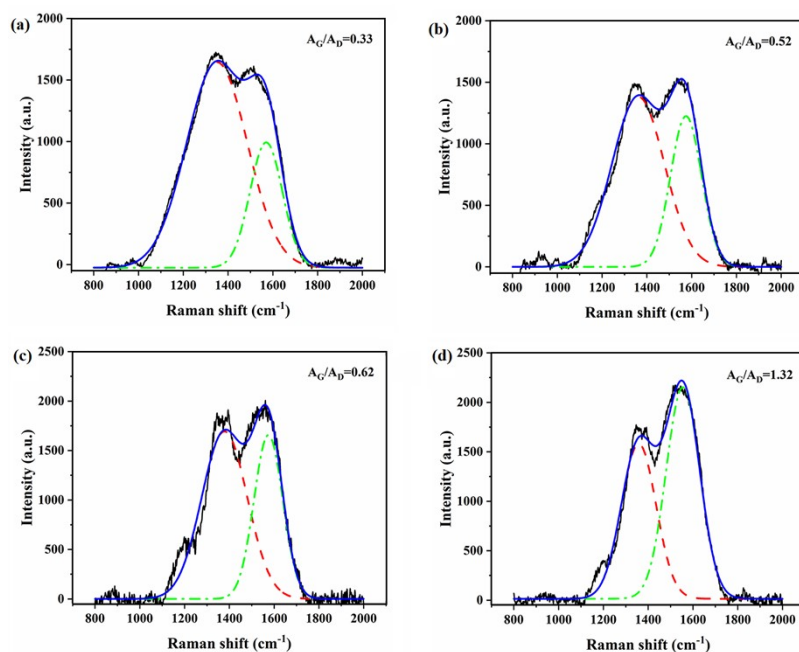
The conversion of the epoxy group ( $\alpha_{\text{epoxy group}}$ ) for CDs/EP composites can be evaluated according to Eq. (2) according to the FTIR spectra. The characteristic peak of the phenyl ring at 1510  $\text{cm}^{-1}$  was selected as the reference.

$$\alpha_{\text{epoxy group}} = 1 - \frac{A / A_{Ph}}{A_0 / A_{0Ph}} \quad (2)$$

where  $A_{0\text{ Ph}}$  and  $A_{Ph}$  are the integrated peak areas of the phenyl ring in polymer systems before and after curing.  $A_0$  and  $A$  are the integrated peak areas of epoxy groups at 910  $\text{cm}^{-1}$  in polymer systems before and after curing.<sup>7,8</sup>

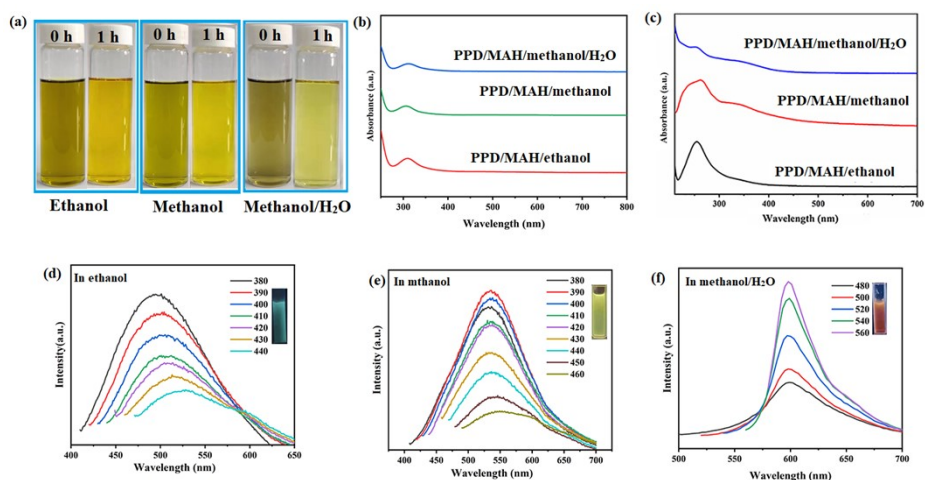


**Fig. S1.** PL spectra of B-CDs (a), G-CDs (b), O-CDs (c) and R-CDs (d) in ethanol. The relationships of integrated emission intensity vs. absorbance and their linear fitting for standard substance and CDs samples (e-h).

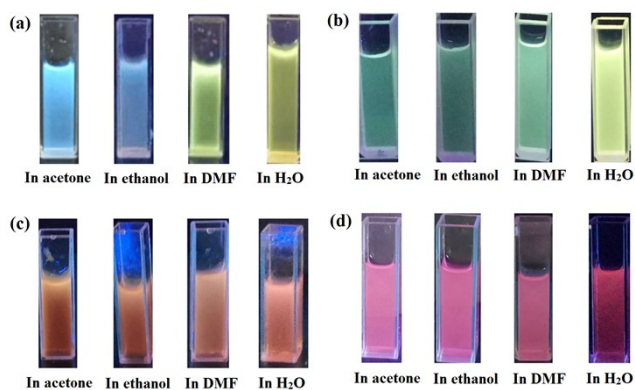


**Fig. S2.** Multiple fitting Raman peaks of B-CDs (a), G-CDs (b), O-CDs (c) and R-CDs (d)

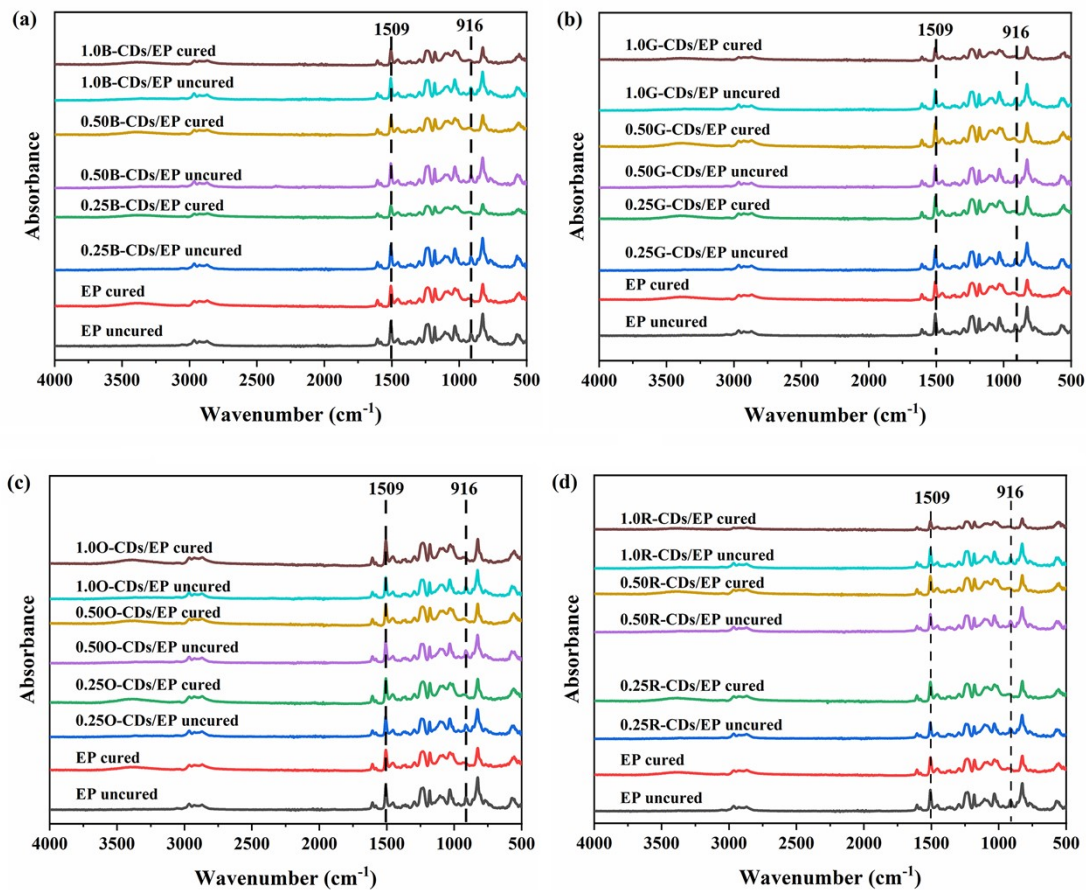
It can be observed that the solutions of PPD/MAH/ethanol, PPD/MAH/methanol and PPD/MAH/methanol/H<sub>2</sub>O become yellow color, light yellow and pale yellow from grey-green color after staying at room temperature for 1h (Fig. S3a), and their UV-vis spectra show that absorbance intensities of the precursors at about 310 nm gradually decrease (Fig. S3b), which indicate that the concentrations of PPD/MAH in ethanol, methanol, and methanol/H<sub>2</sub>O gradually reduce. The UV-vis spectra of the as-prepared CDs in ethanol, methanol and methanol/H<sub>2</sub>O display the significant absorption bands in the region of 200-300 nm, which are attributed to the  $\pi \rightarrow \pi^*$  transition of C=C bonds and C=N in the sp<sup>2</sup> C domain, and they also show weak absorption bands at about 350 nm, corresponding to the  $n \rightarrow \pi^*$  transition of C=O bonds and C=N on the surfaces of CDs (Fig. S3c). The as-prepared CDs in ethanol, methanol and methanol/H<sub>2</sub>O can emit blue, bright yellow-green and red lights under UV irradiation at 365 nm (Fig. 3d-f), and their PL emission wavelengths are at 495-525 nm, 535-555 nm and 600 nm, respectively (Fig. S3d-f). Generally, the emissions of CDs shift toward long-wavelength with increasing solvent polarity. Such change trend is attributed to the fact that with the increase of the solvent polarity from ethanol, to methanol, and then to methanol/H<sub>2</sub>O, the hydrogen bonding interaction between solvent molecules increases, then the hydrogen bonding interaction between solvent and the precursor decreases, which can lead to lower solubility and dispersion of the precursor in solvent. As a results, the prepared CDs display the increased size, which can facilitate the long-wavelength emissions of CDs.



**Fig. S3.** Photographs of raw material in various solvents under ambient light (a). The UV-vis spectra of the raw material in ethanol, methanol and methanol/H<sub>2</sub>O (b). UV-vis spectra of the synthesized CDs in ethanol, methanol and methanol/H<sub>2</sub>O (c). The PL spectra of the synthesized CDs in ethanol (d), methanol (e) and methanol/H<sub>2</sub>O (f) under different excitation wavelengths along with the pictures of CDs solutions under UV irradiation at 365 nm.



**Fig. S4.** Photographs of B-CDs (a), G-CDs (b), O-CDs (c) and R-CDs (d) in acetone, ethanol, DMF and H<sub>2</sub>O under UV light of 365 nm.



**Fig. S5.** FTIR spectra of CDs/EP mixtures before and after curing(a-d).

**Table S1** Fitted parameters of time-resolved PL decay curves and QY of four CDs

Samples	B-CDs	G-CDs	O-CDs	R-CDs
$\lambda_{\text{ex}}$ (nm)	380	390	470	480
$\lambda_{\text{em}}$ (nm)	420	508	596	607
$\tau_1$ (ns)	1.22	1.90	1.83	1.22
$B_1$ (%)	93.87	79.16	33.94	50.63
$\tau_2$ (ns)	13.85	7.71	11.93	8.89
$B_2$ (%)	6.13	20.84	66.06	49.37
$\tau$ (average)	1.99	3.11	8.50	5.01
QY (%)	55.6	76.5	59.5	30.9

**Table S2** Reaction conditions and YQ values of the reported CDs and CDs in this work

Raw materials	Reaction conditions	Relative QY of CDs with different fluorescence /%	Mechanism	Reference
Citric Acid	140-200 °C,	Blue:52.6; Green:35.1	Graphitization	9
Urea	12 h	Red:12.9	-COOH	
Citric Acid	200 °C,	Purple:46.5; Blue:32.3	Graphitization	10
1-(2-pyridylazo)-2-naphthol	7 h	Red:31.6	C=N on surface	
1,3,6 trinitropyrene	230 °C,	Blue:27.5; Green:26.7	Graphitization	6
	12 h	Yellow:71.7; Red:40	Functional groups	
o-phenylenediamine	180 °C,	Blue:4.8; Green:17.6	/	4
m-phenylenediamine	12 h	R:26.1		
p-phenylenediamine				
GO	180 °C,	Blue:18.3; Green:41.4	Graphitization	11
	12 h	Yellow:38.3; Red:18.7; Orange:37.5	Graphite nitrogen	
1,2,4,5-Benzenetetracarboxylic acid	200 °C,	Blue:18; Green:26	Oxygen-containing defects	12
2,7-diaminofluorene	8 h	Yellow: 23, Red:10		
m-phenylenediamine	180 °C,	Blue:25; Green:28	Surface state	13
acid reagents	12 h	YG:72; Red:47		
l-tyrosine	180 °C,	Blue:8.6; Green:12.6	Surface state	14
o-phenylenediamine	18 h	Orange:20.9		
p-phenylenediamine	180 °C	Blue:61.6; Green:41.3	O-related and N-related defect states	15
	12 h	Red:29.1		
Catechol	200 °C,	Blue:50.54;	Graphitization	16
m-dihydroxybenzene	2-7 h	Green:41.21	Oxygen element	
hydroquinone		Yellow:38.8		
1,6-dihydroxynaphthalene		Orange:36.92		
		Red:23.85		
p-phenylenediamine	180 °C,	Blue:55.6; Green:76.5	Graphitization	This work
maleic anhydride	4 h	Orange:59.5; Red:30.9	Functional groups	

**Table S3** Properties of the reported CDs/EP and CDs/EP in this work

Content of CDs (wt%)	Tensile Strength (MPa)	T <sub>di</sub> (°C)	Reference
0.1-1.0	40.7-46	/	17
1.0	62.5	291	18
1.0-5.0	4.6-7.8	/	19
1.0-10.0	28.1	/	20
0.25-1.0	60-69	321-332	This work

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