## Supplementary Materials

## **Preparation of Multicolor Photoluminescent Carbon Dots by Tuning Surface States**

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**Figure S1** a) PL emission spectra of the g-CDs ethanol dispersion under different excitation wavelengths and PL excitation spectrum at emission wavelength of 510 nm; b) PL emission spectra of the r-CDs ethanol dispersion under different excitation wavelengths and PL excitation spectra at emission wavelengths of 600, 653 and 720 nm.



**Figure S2** a) UV-Vis absorption and PL excitation spectra of b-CDs and g-CDs (excitation wavelengths at 435 and 510 nm, respectively); b) UV-Vis absorption and PL excitation spectra of y-CDs and r-CDs (excitation wavelengths at 535 and 650 nm, respectively).



**Figure S3 a)** PL decay spectra of the g-CDs ethanol dispersion monitored at 510 nm under excitation of 457 nm; b) PL decay spectra of the r-CDs ethanol dispersion monitored at 602, 650 and 714 nm under excitation of 588 nm.



**Figure S4** a) XPS survey of the b-CDs, g-CDs, y-CDs, and r-CDs; b) Relative contents of C, N, and O elements of these CDs based XPS data.



**Figure S5** a-b) high resolution XPS N 1s spectra of g-CDs and r-CDs and their comparison to b-CDs and y-CDs, respectively; c-d) high resolution XPS O 1s spectra of g-CDs and r-CDs and their comparison to b-CDs and y-CDs, respectively.



**Figure S6** Photographs of the b-CDs@PVP, g-CDs@PVP, y-CDs@PVP, and r-CDs@PVP powders (from left to right) under daylight and UV light (365 nm).



**Figure S7** PL emission spectra under different wavelengths and excitation spectra at the corresponding emission maxima of b-CDs@PVP (a), g-CDs@PVP (b), y-CDs@PVP (c), and r-CDs@PVP (d) powders.



Figure S8 Corresponding emission spectra of the multicolour LEDs.

Sample	λ <sub>ex</sub> (nm)	<b>Φ</b> <sub>1</sub> (%)	<b>\$\$</b> 2(%)	<b>Ф</b> <sub>3</sub> (%)	<b>Φ</b> _4(%)	<b>Φ</b> <sub>5</sub> (%)	<b>Ф<sub>аvg</sub>(%)</b>	<b>Φ</b> <sub>corr</sub> (%)
R-6G	488	93.45	94.33	93.18	94.02	94.27	93.85	95
g-CDs	440	28.21	28.02	27.91	27.75	27.50	27.88	28.22
r-CDs	540	21.09	23.74	21.47	21.14	21.30	21.75	22.01

Table S1. QYs of the g-CDs and r-CDs in ethanol.

Table S2. Fitted parameters of the PL decay spectra of the g-CDs and r-CDs.

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Sample	λ <sub>ex</sub> (nm)	λ <sub>em</sub> (nm)	τ₁(ns)	B₁(%)	φ
g-CDs	457	510	4.86	100	1.067
r-CDs	588	608	2.18	100	1.096
r-CDs	588	650	2.20	100	1.133
r-CDs	588	714	2.24	100	1.057

Table S3. PL QYs of the b-CDs@PVP, g-CDs@PVP, y-CDs@PVP and r-CDs@PVP powders.

Sample	$\lambda_{ex}$ (nm)	$\boldsymbol{\Phi}_1$	$\boldsymbol{\phi}_2$	${oldsymbol{\phi}}_3$	$oldsymbol{\Phi}_4$	$oldsymbol{\phi}_{5}$	$oldsymbol{\phi}_{avg}$
b-CDs-PVP	360	9.42	8.86	9.01	8.04	8.77	8.82
g-CDs-PVP	450	15.36	14.93	14.74	14.03	13.50	14.44
y-CDs-PVP	430	16.19	16.06	16.25	15.97	16.18	16.13
r-CDs-PVP	540	15.74	14.94	14.78	14.39	14.20	14.81