Supporting Information

Carbon dots/ZnO quantum dots composite based white phosphors for white light-emitting diodes

Wenjing Li[a,#], Min Wu[b,#], Hang Jiang[a], Longhui Yang[a], Chao Liu[a], Xiao Gong*[a]

a. State Key Laboratory of Silicate Materials for Architectures, Wuhan University of Technology, Wuhan 430070, P. R. China.

b. Huaxi MR Research Center (HMRRC), Department of Radiology, Functional and Molecular Imaging Key Laboratory of Sichuan Province, West China Hospital, Sichuan University, Chengdu, 610041, China

# equal contribution

* Corresponding author.

E-mail address: xgong@whut.edu.cn (X. Gong)

Experimental

Chemicals and materials

Zinc acetate dihydrate ($Zn(Ac)_2·2H_2O$, 99%), o-phenylenediamine (98%), L-ascorbic acid (99%), (3-Aminopropyl)triethoxysilane (APTES, 98%) were purchased from Aladdin. Potassium hydroxide (KOH, 98%), ethanol (99.9%), polyvinyl pyrrolidone (PVP) were supplied by Sinopharm Chemical Reagent. Epoxy resins (E51 + W93) were purchased from Kunshan Jiulimei Electronic Materials Co., Ltd. The chemical reagents were used as received without further purification. 365 nm LED chips were purchased from Looking Long Technology Co., Ltd.

Preparation of B-CDs/ ZnO QDs composites

Preparation of B-CDs:

0.2 g o-phenylenediamine and 0.5 g L-ascorbic acid were mixed with 20 mL ethanol in a 50 mL beaker. After ultrasonic, the mixed solution was transferred to a 25 mL polytetrafluoroethylene (PTFE)-lined reaction kettle in an oven at 180 ℃ for 6 h And after being cooled naturally to room temperature, the solution was collected and was centrifuged at 8000 r/min for 10 min. The upper clarified solution was filtered using a 0.22 μm filter membrane to remove large particulate matter, and the clear solution was obtained.

Preparation of ZnO QDs:

1.1 g $Zn(Ac)_2·2H_2O$ were dissolved in 30 mL ethanol and the mixture was continuously stirred vigorously until it was clear and transparent at 80 ℃ in a three-

necked flask. The solution was chilled to 50 ℃ by the ice and water mixture. And then 4 mL KOH ethanol solution (1.75 M) was injected into the above solution. The reaction proceeded for 2 h. To increase the water dispersion of ZnO QDs, 1 mL APTES was dispersed in 4 mL deionized water, ultrasonic dispersion lasted for 5 minutes, and then injected into the above reaction solution. Under this condition, the reaction continued for 1 h, and white precipitate would appear in the reaction. After the reaction, the solution was centrifuged at 8000 r/min for 10 min, and the white precipitate was collected. The white precipitate was washed by ethanol 3 times. The obtained precipitate was dried in an oven at 80 ℃.

Preparation of B-CDs/ZnO QDs composites:

10 mL B-CDs solution was added into a 100 mL beaker and was continuously stirred vigorously for 1 h. 0.01 g, 0.02 g, 0.03 g, 0.04 g of water-dispersible ZnO QDs were respectively and slowly added to the continuously stirred B-CDs solution and stirred for 6 h. Then, 0.8 g PVP was slowly added to the above solution. The mixture was dried at 40 ℃ for 48 h and got the solid sample.

Preparation of white LED devices

An appropriate amount of the composite powder and epoxy resin A glue was mixed evenly. At the same time, adding matching curing agent B according to the A, B glue ratio 10:3, and then stirring until evenly dispersed. The evenly mixed sample was dropped on the 365 nm chip and cured at room temperature. Furthermore, ZnO QDs monochrome yellow LED was prepared by mixing ZnO QDs powder with epoxy resin

A and B using the same encapsulation method.

Characterization

Zeta potential was measured by Malvern (UK) Zetasizer Nano ZS. Transmission electron microscopy (TEM) characterization was carried out using a JEM-2100F TEM (JEOL, Japan). Ultraviolet-visible (UV–vis) absorption spectra were recorded with a UV–Vis spectrophotometer (UV2600, Shimadzu Corp., Tokyo, Japan) in the range of 250-800 nm. Photoluminescence (PL) spectra were measured at room temperature with a time-resolved fluorescence spectrometer (FL3-22, Jobin-Yvon, USA). The fluorescence lifetimes were measured using a time-correlated single-photon counting (TCSPC) spectrometer (Newport, USA) with a femtosecond laser (Spirit 1040-8W, Spectra-Physics, USA). The X-ray diffraction (XRD) was performed using a D8 Advance diffractometer (Bruker, Germany, K_{α}=1.5406 Å). Fourier transform infrared spectroscopy (FT-IR) spectra of samples were recorded from 400-4000 cm^{-1} using Nicolet 6700. The X-ray photoelectron spectroscopy (XPS) spectra of the samples were collected by using a Thermo Fisher Scientific ESCALAB 250Xi spectrometer equipped (USA). The electroluminescence (EL) spectra of WLEDs were measured using an HP9000 LED fast spectrum analyzer. The absolute photoluminescence quantum yield (PLQY) was measured using EI FLS1000 Photoluminescence Spectrometer.

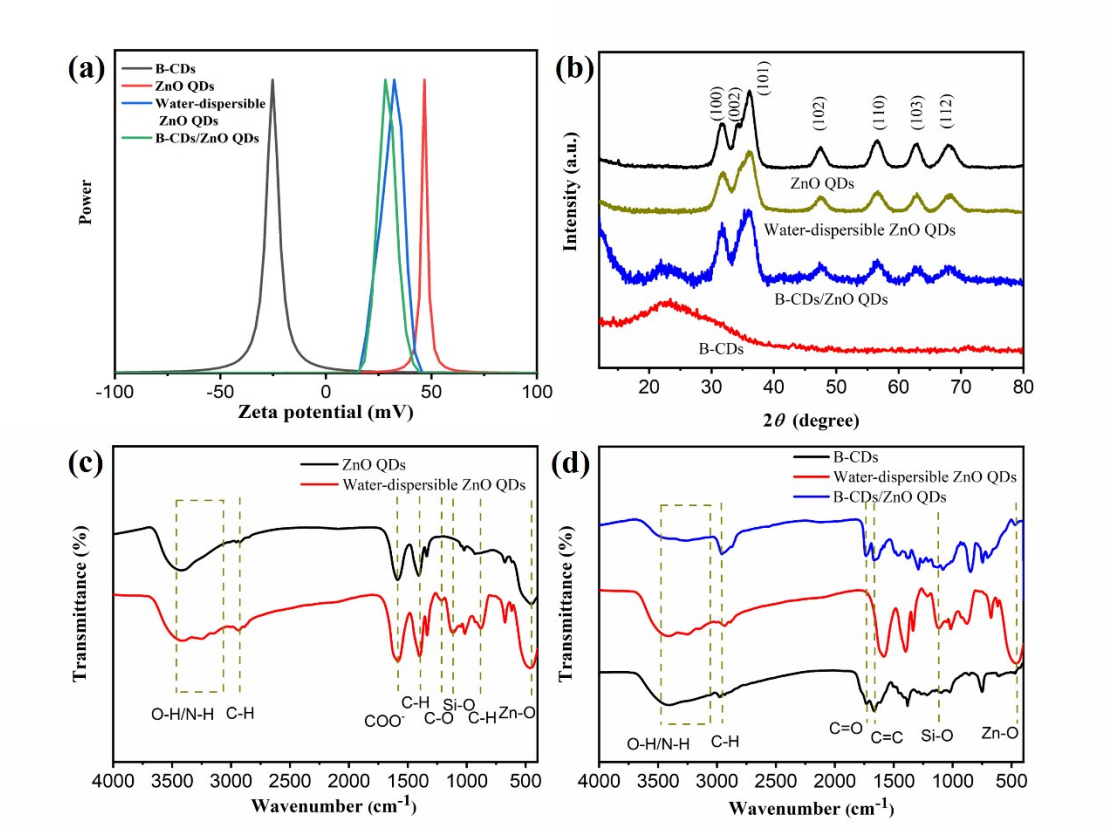

Fig. S1 (a) Zeta potential of B-CDs, ZnO QDs, water-dispersible ZnO QDs, and B-CDs/ZnO QDs. (b) XRD patterns of B-CDs (red line), B-CDs/ZnO QDs (blue line), water-dispersible ZnO QDs (yellow line), ZnO QDs (black line), and the standard XRD pdf card of ZnO; FT-IR spectra of (c) ZnO QDs and water-dispersible ZnO QDs, (d) B-CDs, water-dispersible ZnO QDs, and B-CDs/ZnO QDs.

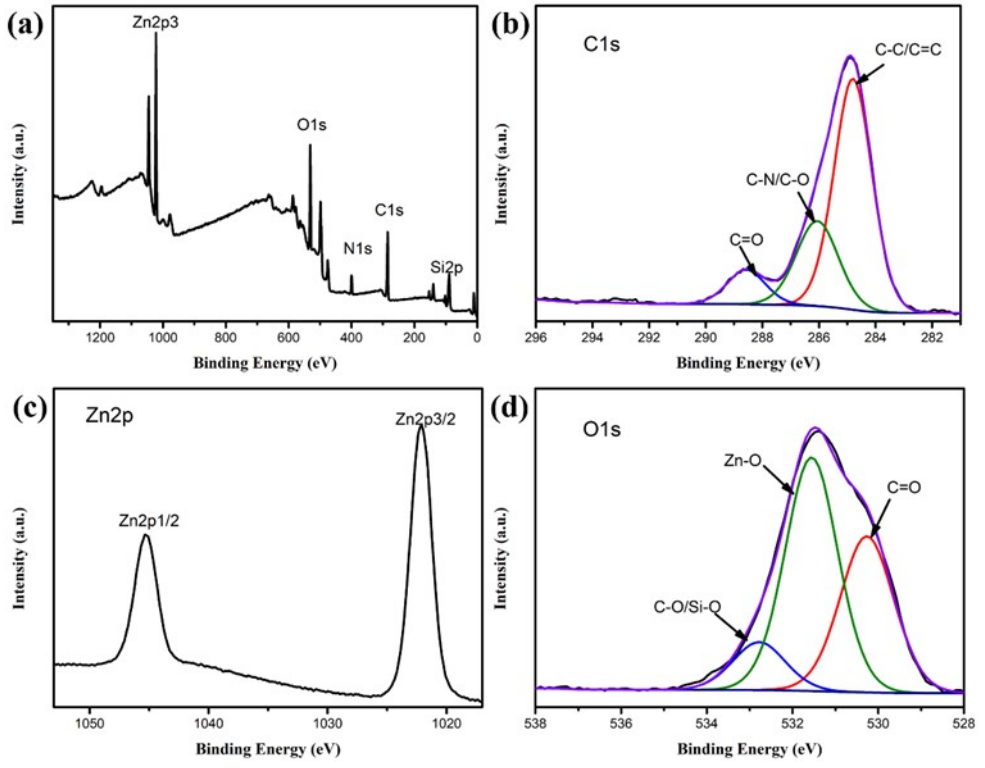

Fig. S2 Survey XPS spectrum of B-CDs/ZnO QDs. High-resolution XPS spectrum of (b) C1s (c) Zn2p (d) O1s.

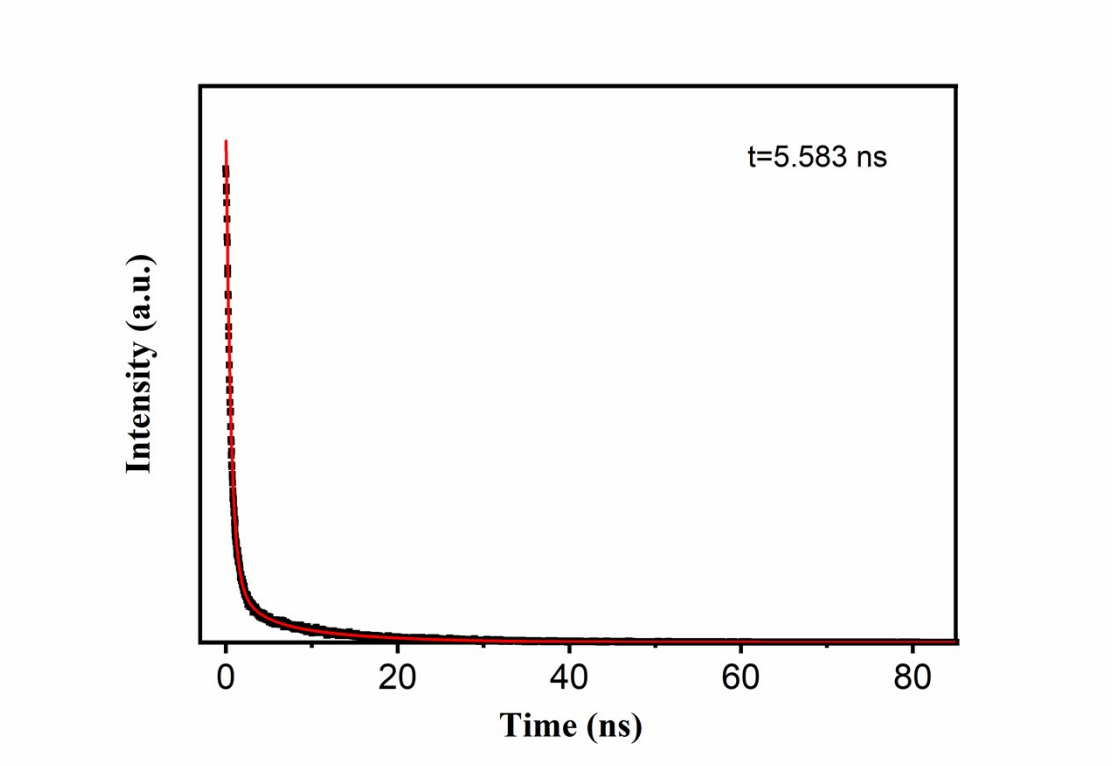

Fig. S3 Fluorescence decay curvy of B-CDs aqueous solution (λ_{ex} = 375 nm, λ_{em} = 445 nm).

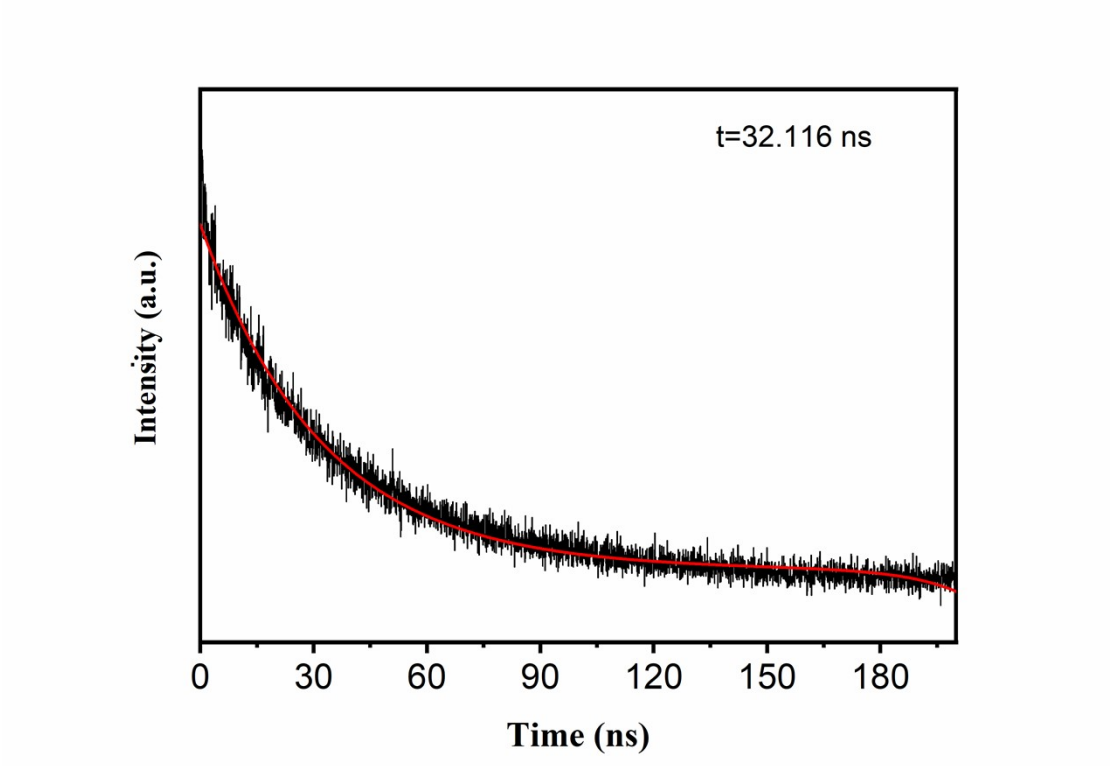

Fig. S4 Fluorescence decay curvy of ZnO QDs aqueous solution (λ_{ex} = 365 nm, λ_{em} = 550 nm).

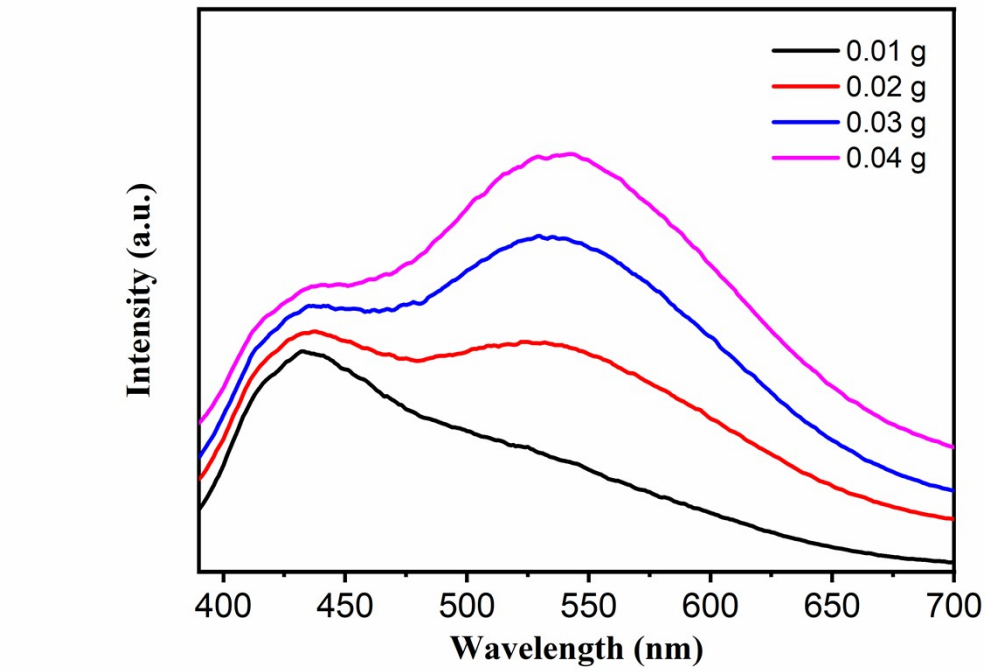

Fig. S5 Photoluminescence emission spectra of B-CDs/ZnO QDs composite materials with different ZnO QDs loadings (λ_{ex} = 365 nm).

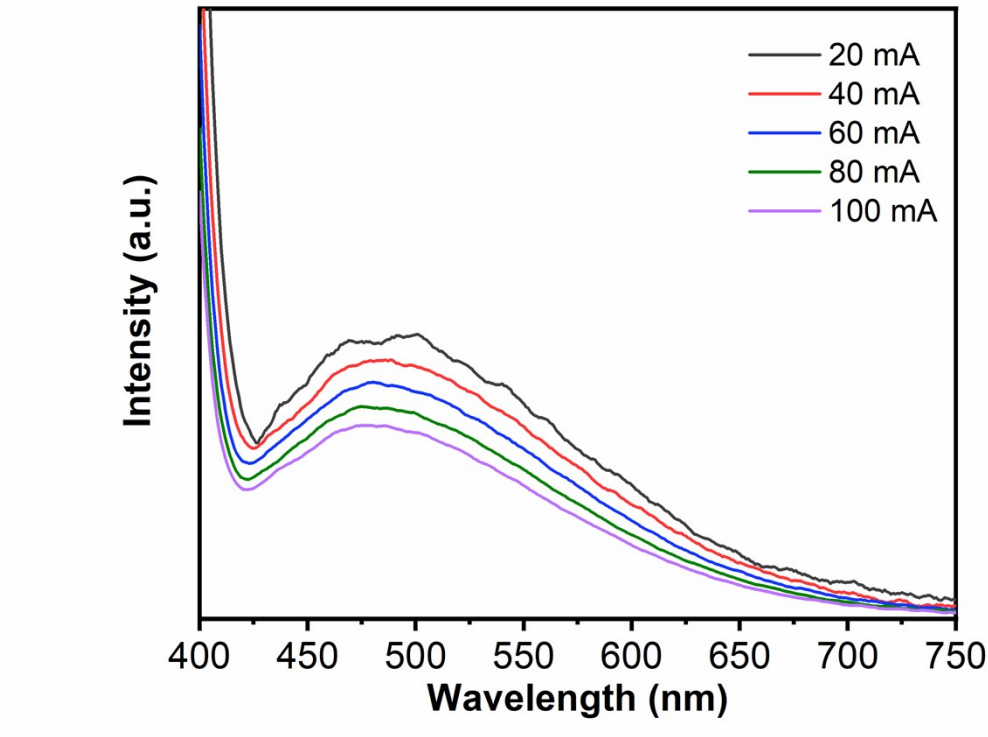

Fig. S6 Luminescence emission spectra of WLEDs under different currents.

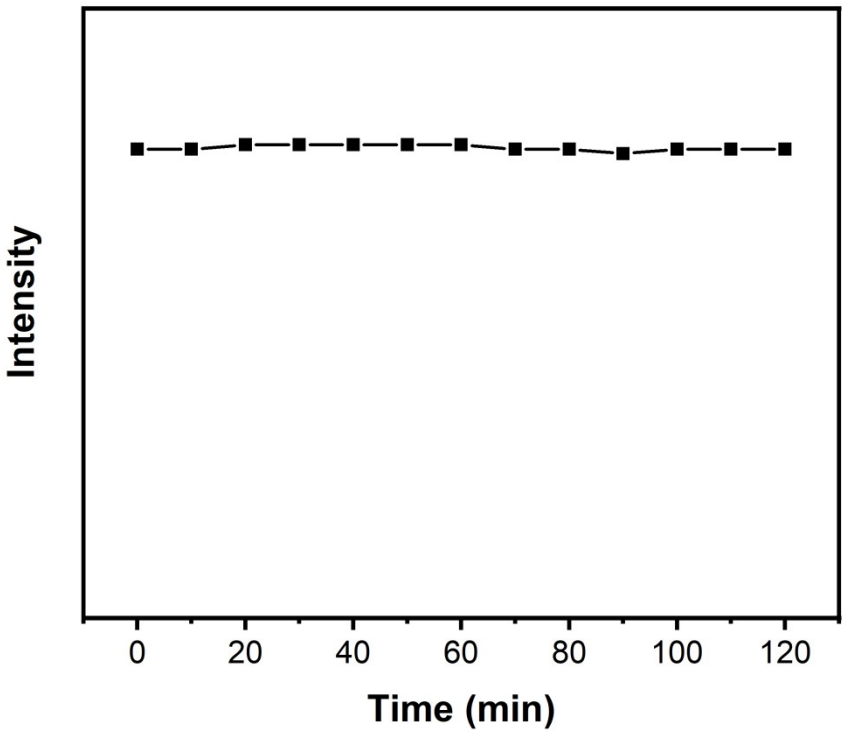

Fig. S7 Luminescence intensity of WLEDs under different working time.

Table S1 The PLQY of CDs, ZnO QDs and the composites in solution and solid state.

Materials	PLQY
CDs (solution)	4.55 %
ZnO QDs (solution)	24.92 %
CD/ZnO QDs (solution)	7.92 %
CD/ZnO QDs (solid)	3.00 %

Table S2 The CIE coordinates, color temperature and luminous efficiency of the LED devices with different amount of ZnO QDs.

Amount of ZnO QDs	CIE coordinates	Color temperature (K)	Luminous efficiency (lm/W)
0.01 g	(0.249, 0.313)	11383	2.38
0.02 g	(0.255, 0.319)	10816	2.80
0.03 g	(0.264, 0.332)	9534	2.85