Supporting Information

Matrix-Free Thermally Activated Delayed Fluorescent Carbon Dots-based Electroluminescent Light-Emitting Diodes Exceeding 5.6 % External Quantum Efficiency


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**Experimental Procedures**

**Materials:** Pyrene formaldehyde and diaminobenzophenone were purchased from Sigma-Aldrich Co., Ltd. PEDOT:PSS, PVK, NPB, m-MTDATA, TPBi, LiF, Al were purchased from Xi'an Yuri Solar Co., Ltd. All the reagents were analytical grade and utilized without further purification. The redistilled water used in these experiments was purified using a SZ-93A water purification system.

**Preparation of Carbon Dots (CDs):** CDs were synthesized by a solvothermal method. Briefly, pyrene formaldehyde (2 mmol), diaminobenzophenone (1 mmol) and sulfuric acid (500 µL) were dissolved in ethanol (10 mL), stirred for 15 min, then the solution transferred to a 25 mL poly (tetrafluoroethylene)-lined autoclave and heated at 200 °C for 6 h. After cooling to room temperature, the solution was centrifuged at 10000 rpm for 5 min, filtrated through a 0.22 μm filter membrane, and finally dialyzed in ethanol with the membrane (1000 Da) for 1 day to remove any unreacted precursor and acid., then the pure CDs was obtained by further column chromatography using mixture of dichloromethane and ethyl acetate as the eluent.

**Characterization of CDs:** Transmission electron microscopy (TEM) images were acquired with a FEI TECNAI G2F20-S-TWIN electron microscope. X-ray photoelectron spectroscopy (XPS) measurements used a Thermo Fisher ESCALAB 250Xi surface analysis system. X-ray diffraction (XRD) patterns were obtained using an X-ray diffractometer (PANalytical, X’Pert PRO). The absorption and fluorescence spectra of the CDs were recorded on a Persee TU-1810PC spectrophotometer and FLS1000 fluorescence spectrophotometer, respectively, at room temperature. Fourier transform infrared spectroscopy (FTIR) data were collected on a Nexus 470 (Thermo Fisher) spectrometer. The thickness of each layer of electroluminescent LEDs was obtained by Dektak XT probe profilometer (Bruker).

**Device fabrication and characterizations:** Clean ITO substrates were firstly washed with ethanol and then dried under a stream of flowing N₂ gas. Next, the ITO substrates were treated by ultraviolet ozone for 15 min. Subsequently, PEDOT: PSS was spin-coated onto the clean ITO substrates at 4000 rpm. for 40 s, followed by annealing at 140 °C for 10 min. The obtained ITO/PEDOT: PSS films were then spin-coated with a PVK solution (8 mg/mL in chlorobenzene) at 3000 rpm. for 40 s, followed by thermal treatment at 140 °C for 15 min. Then, a CDs solution (8 mg/mL m-MTDATA: 8 mg/mL CDs) was spin-coated onto the films at 2000 rpm. for 40 s, followed by thermal treatment at 90 °C for 15 min. Finally, the films were transferred to an evaporation chamber, and coated successively with 40 nm TPBi, 1 nm LiF, and 80 nm Al. The device area was 9 mm². The current-voltage characteristics, electroluminescence spectra, luminance, external quantum efficiency, and operation lifetime of the devices were measured using a computer-controlled Integrated system, which is consists of a Keithley 2400 source meter, a fiber integration sphere (FOIS-1) along with a spectrometer.
Results and Discussion

Figure S1. Height analysis of CDs

![Figure S1](image1.jpg)

Figure S2. DTG analysis of CDs.

![Figure S2](image2.jpg)

Figure S3. XPS (a) C 1s, (b) N 1s, and (c) O 1s spectra of CDs.

![Figure S3](image3.jpg)
Figure S4. PLQY of CDs solution.

Figure S5. Fluorescence lifetime of CDs solution.
Figure S6. TADF lifetime of CDs solution.

Figure S7. TRES of CDs solution.
Figure S8. TRES of the prompt and delayed emission of CDs solution.

Figure S9. Quantum yield of CDs films with different ratios of CDs.
Figure S10. Thickness analysis of each layer of electroluminescent LEDs

Figure S11. The CIE color coordinates of Y-LEDs and W-LEDs.

Figure S12. Normalized EL spectra of (a) Y-LEDs and (b) W-LEDs.
**Figure S13.** Current density-luminance-voltage (J–L–V) characteristic curves for (a) Y-LEDs and (b) W-LEDs.

**Figure S14.** Current density-Current efficiency (J–C.E.) characteristic curves for (a) Y-LEDs and (b) W-LEDs.

**Figure S15.** EQE-J curves of (a) Y-CDs with different thicknesses of TPBi and (b) W-LEDs with different thicknesses of emission layer.