

## SUPPORTING INFORMATION

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### Supporting Information

#### **Temperature-Responsive Conversion of Thermal Activated Delayed Fluorescence and Room Temperature Phosphorescence of Carbon Dots in Silica**

Yuqiong Sun, Jinkun Liu, Xiaoliang Pang, Xuejie Zhang, Jianle Zhuang, Haoran Zhang, Chaofan Hu, Mingtao Zheng\*, Bingfu Lei\*, Yingliang Liu\*

### Experimental section

*Chemicals and Materials:* The rice husk (RH) were collected from a mill in Guangzhou (China). Sodium hydroxide (NaOH), diethylenetriamine (DETA) and ethylene glycol were (EG) were bought from Aladdin Chemicals Co. Ltd (Shanghai, China). Hydrochloric acid (HCl) was purchased from Guangzhou Chemical Reagent Co., Ltd (China). Ethylenediamine (EDA), ethanolamine (ETA), and citric acid anhydrous (CA) were obtained from Macklin Chemicals Co. Ltd (Shanghai, China). All the chemicals in this work were used as received without further purification.

*Preparation of p-CDs@SiO<sub>2</sub> gel:* 10 g RH was ground into powder and soaked in 2 M HCl solution for 2 h, and then dried after washed with deionized water to neutral. The pretreated RH were first dispersed in 100 mL of NaOH solution (0.8 M) and mixed with 6 ml EDA (or 2 ml, 4 ml, 8 ml, 10 ml) in a round bottom flask with magnetic stirring at 180 °C for 6 h. Then, the mixture CDs-contained liquid was obtained after filtered (and free CDs were obtained from purified mixture liquid for further use), HCl (3M) was dropwise into the above mother liquor until the pH is equal to 9 to ensure proper gelation. The as-resulted solution was then aged for 5-8 h to form a chocolate-brown gel, which was then washed with deionized water to remove surface-attached CDs and inorganic salts. Finally, the gel was dried at 60 °C for 10 h in an oven, and the faint yellow p-CDs@SiO<sub>2</sub> gel were prepared.

As for other EDA analogues (DETA, ETA, EG, CA), the experiments were carried on under the same identical conditions, except that the EDA was replaced with analogues. Among them, the usage of solid analogues was 1 g.

*Preparation of CDs@SiO<sub>2</sub> composite:* The CDs@SiO<sub>2</sub> composite was obtained by calcine above p-CDs@SiO<sub>2</sub> gel in the muffle furnace. During the calcination process, a ramp rate of 5 °C /min and temperature range of 400-700 °C was used to processing samples, after calcined for 1h in air, the white CDs@SiO<sub>2</sub> composite was obtained.

*Preparation of CD1@CaF<sub>2</sub> composite:* CD1@CaF<sub>2</sub> composite was prepared according to a reported method. <sup>[S1]</sup> Briefly, 1 g of citric acid, 30 mL of deionized water, and 1.6 mL of ethylenediamine was heated at 200 °C for 5 h via a hydrothermal method and then cooled down to room temperature, the CD1 were obtained. The CD1 were added into ammonium fluoride and stirred for 15 min; anhydrous calcium chloride dissolved in 20 mL deionized water was slowly dropped into above solution within 15 min and continue stirring for 1 h. Finally, the solid CD1@CaF<sub>2</sub> product was obtained by suction filtration and dry.

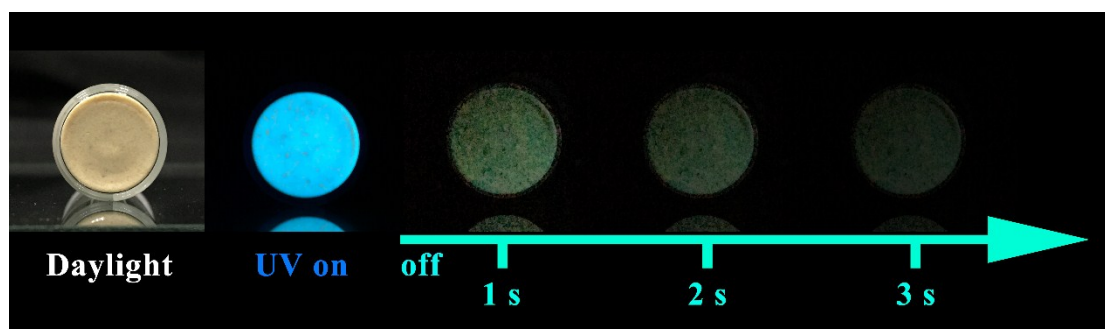
*Visualization of invisible fingerprints:* To detect the invisible fingerprints, the hands should be

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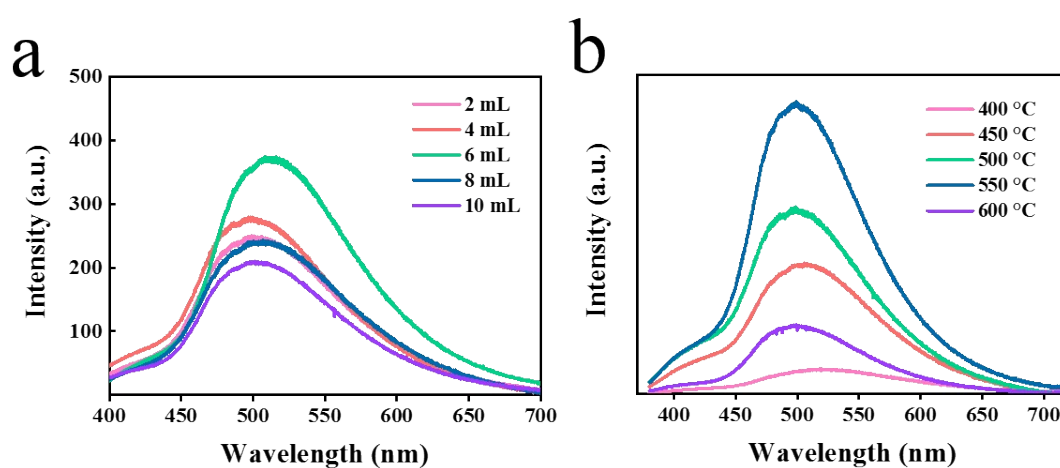
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washed first, then rubbed fingers on the forehead and pressed on the surface of different substrates (weighing paper, glass, metal and plastics). Subsequently, the CDs@SiO<sub>2</sub> powder was spreaded evenly on the surface and hold for a few seconds, the phosphorescent fingerprint was obtained after remove excess powder by a squirrel-hair brush carefully. The phosphorescent images were recorded after 365 nm UV excitation using a mobile phone.

*Characterization:* Transmission electron microscope (TEM) images were carried out on JEOL-2010. Powder X-ray diffraction (XRD, Rigaku) were measured in the 2θ range from 10° to 80° in order to grasp the structures of the samples. The infrared spectrum was recorded by a Nicolet Avatar 360 Fourier transform-infrared spectroscopy (FTIR) from 500 to 4000cm<sup>-1</sup> wavenumber using KBr as a reference to analyze the sample chemical composition. The X-ray photoelectron spectroscopy (XPS) experiments were characterized using a Thermo Fisher 250Xi X-ray photoelectron spectrometer with a monochromatic Al Ka X-ray source. The UV-Vis absorption spectra of the samples were acquired on a Shimadzu UV-2550 ultraviolet-visible spectrophotometer. Photoluminescence (PL) spectra and time-resolved decay curve were measured with Hitachi Model F-7000 fluorescence spectrophotometer equipped with the 150W Xenon lamp as the excitation source. Temperature-dependent spectra were recorded with an Edinburgh S1000 fluorospectrophotometer. The optical microscope photographs were obtained using an optical microscope.



**Figure S1.** Photographs of p-CDs@SiO<sub>2</sub> gel under sunlight, excited with 365 nm UV lamp, and after switching off UV lamp.



**Figure S2.** (a) Phosphorescence emission spectra of CDs@SiO<sub>2</sub> composite obtained by adding different amount of EDA. (b) Phosphorescence emission spectra of CDs@SiO<sub>2</sub> composite calcined at different temperature.

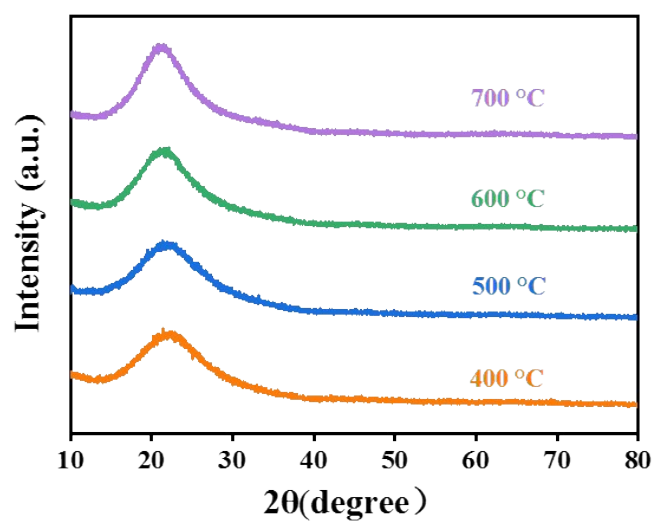


Figure S3. XRD patterns of CDs@SiO<sub>2</sub> composite calcined at different temperature.

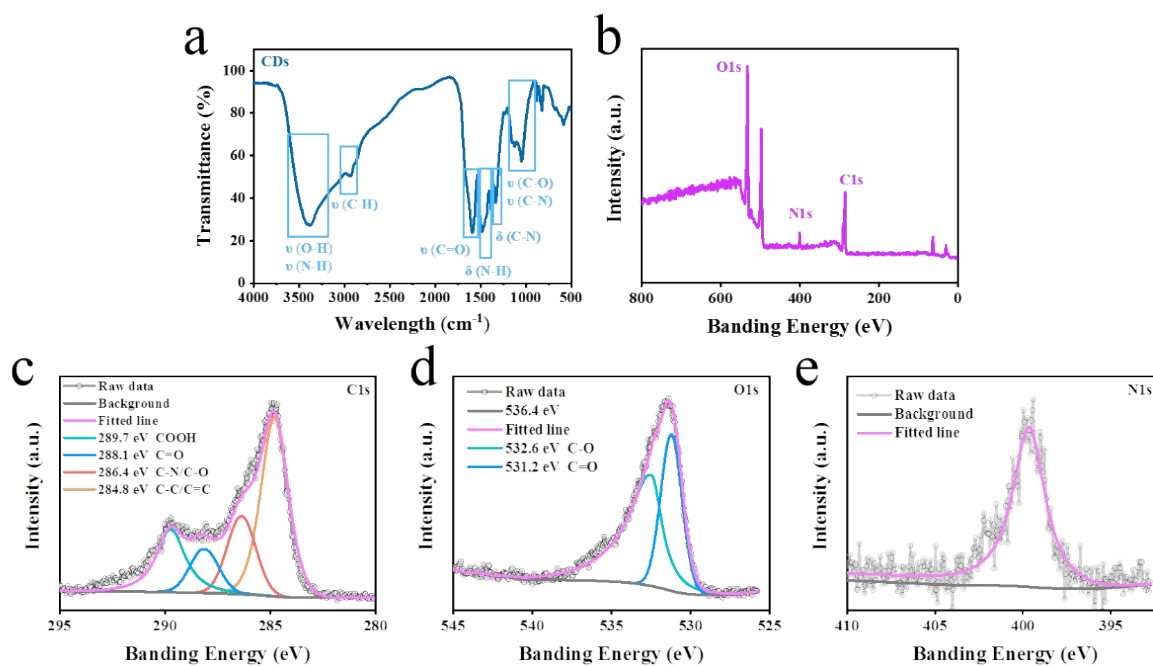
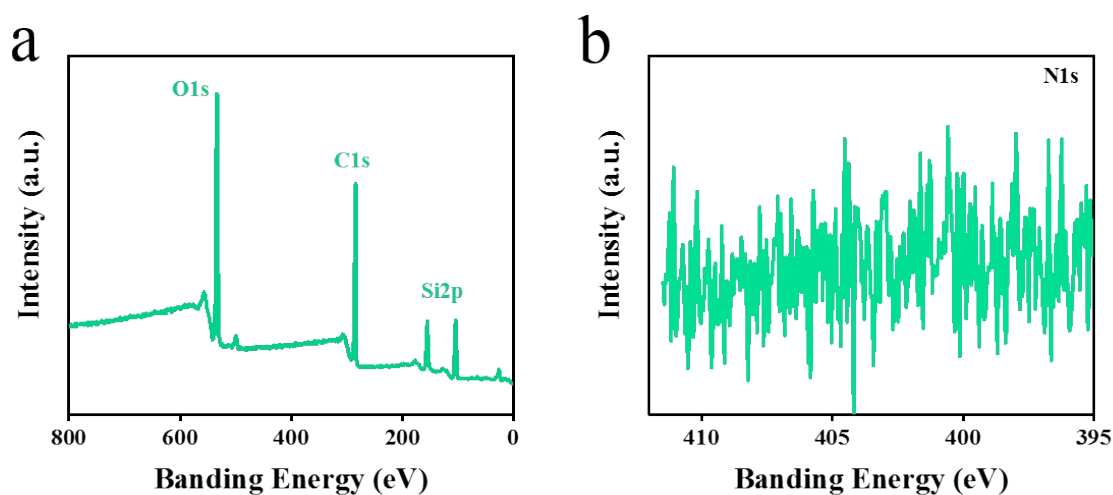


Figure S4. (a) FTIR spectra of CDs. (b) XPS survey spectrum of CDs. (c-e) High resolution XPS

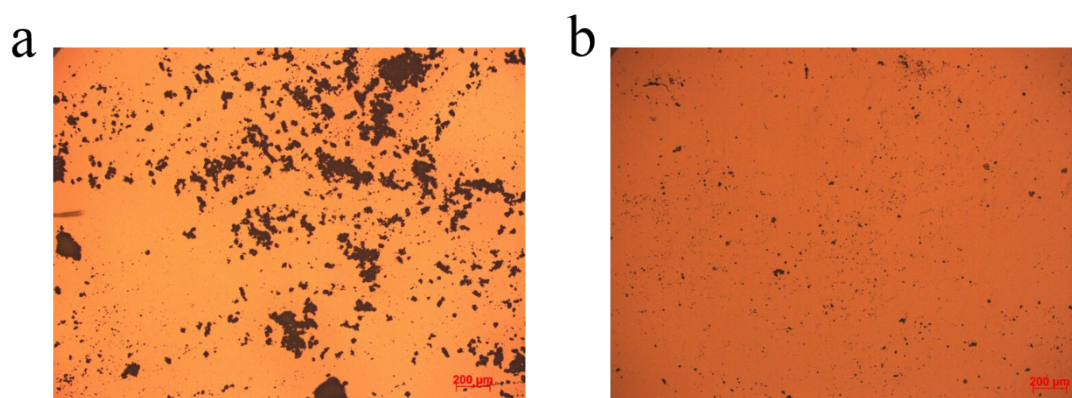
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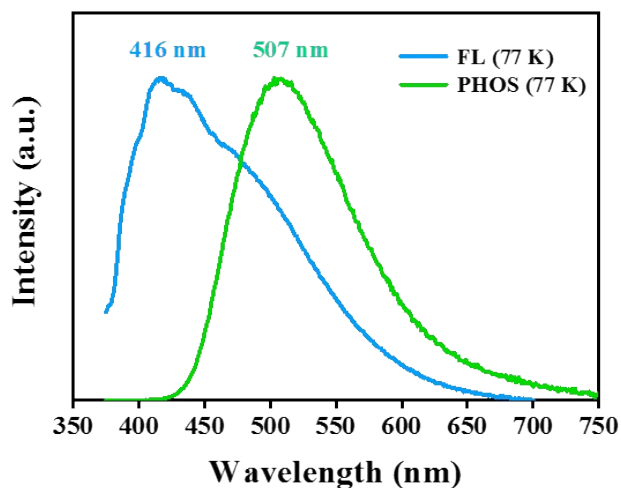
spectrum and fitting results of C 1s (c), O 1s (d) and N 1s (e) of CDs.



**Figure S5.** (a) XPS survey spectrum of CDs@SiO<sub>2</sub>. (b) High resolution XPS spectrum of N1s of CDs@SiO<sub>2</sub>.

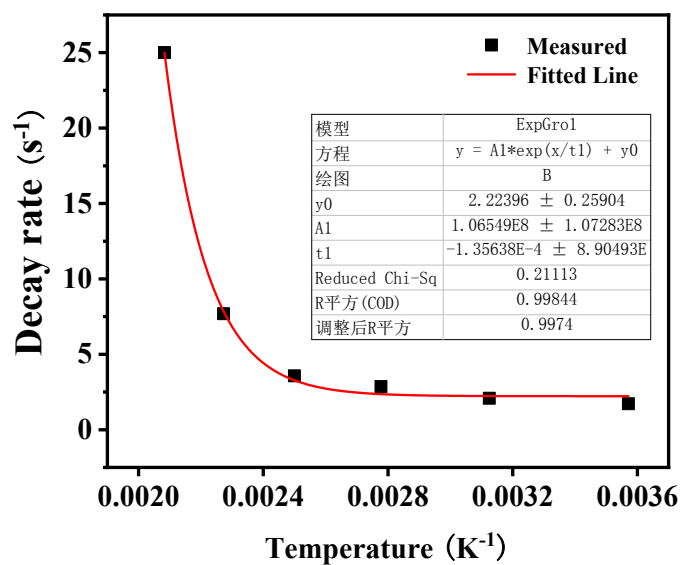


**Figure S6.** The optical microscope photographs of ground CDs@SiO<sub>2</sub> powder (a) and its dispersion liquid (b).



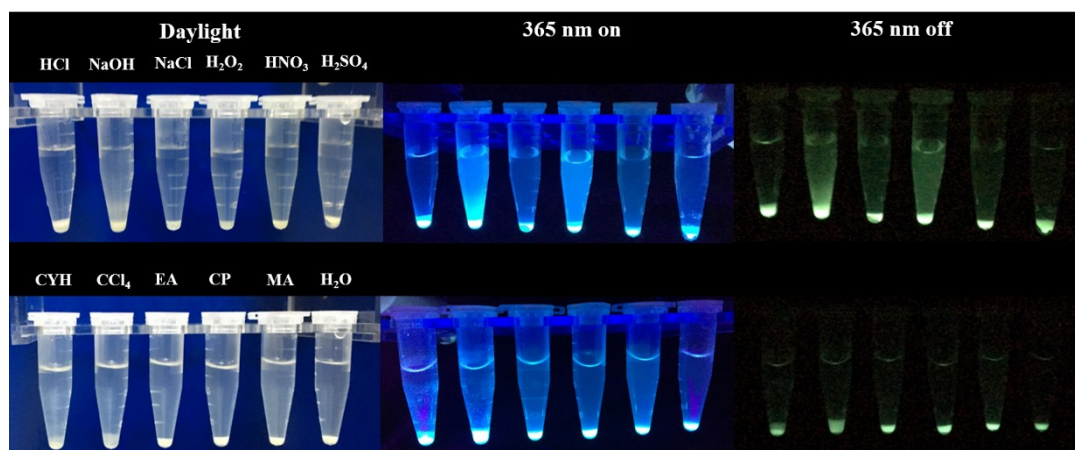
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**Figure S7.** Fluorescence and phosphorescence emission spectra of CDs@SiO<sub>2</sub> at 77 K.

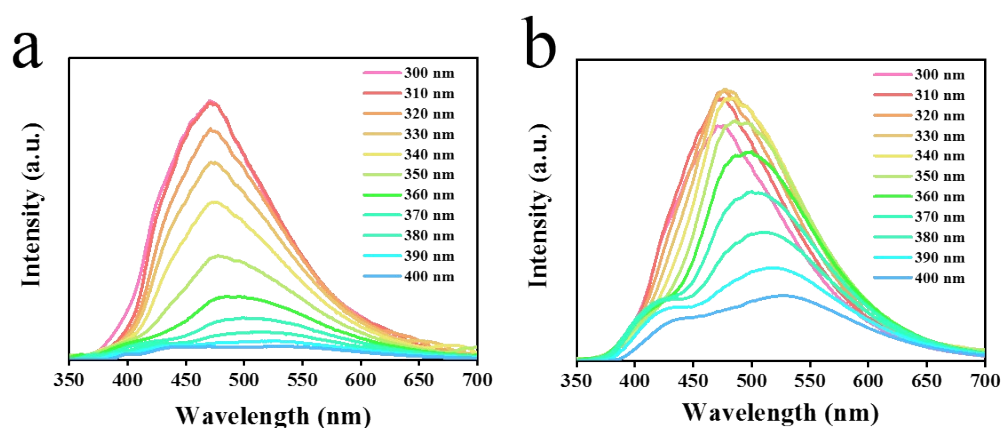


**Figure S8.** Afterglow decay rates of CDs@SiO<sub>2</sub> as a function of temperature and fitting curve based on eq 2.

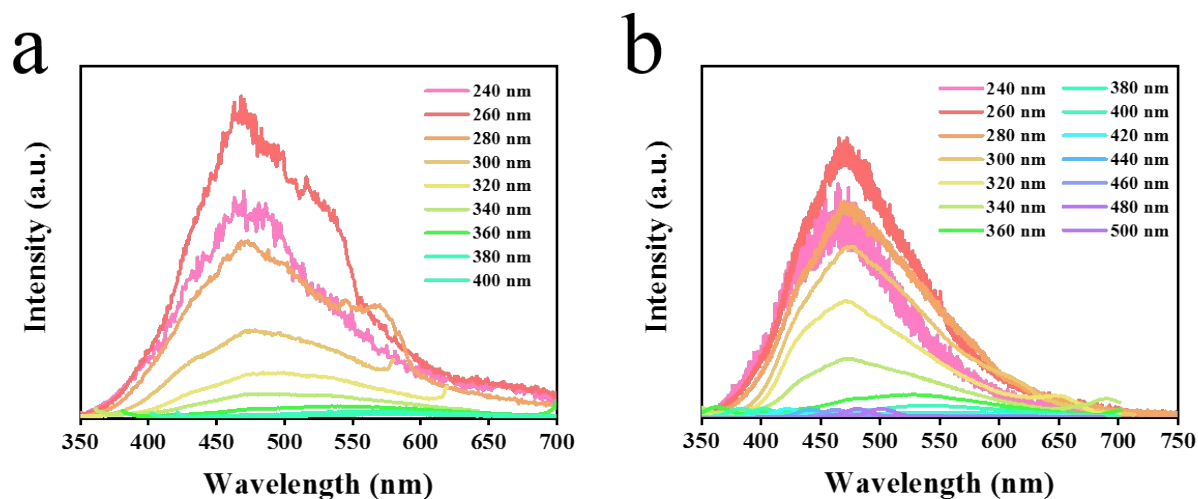




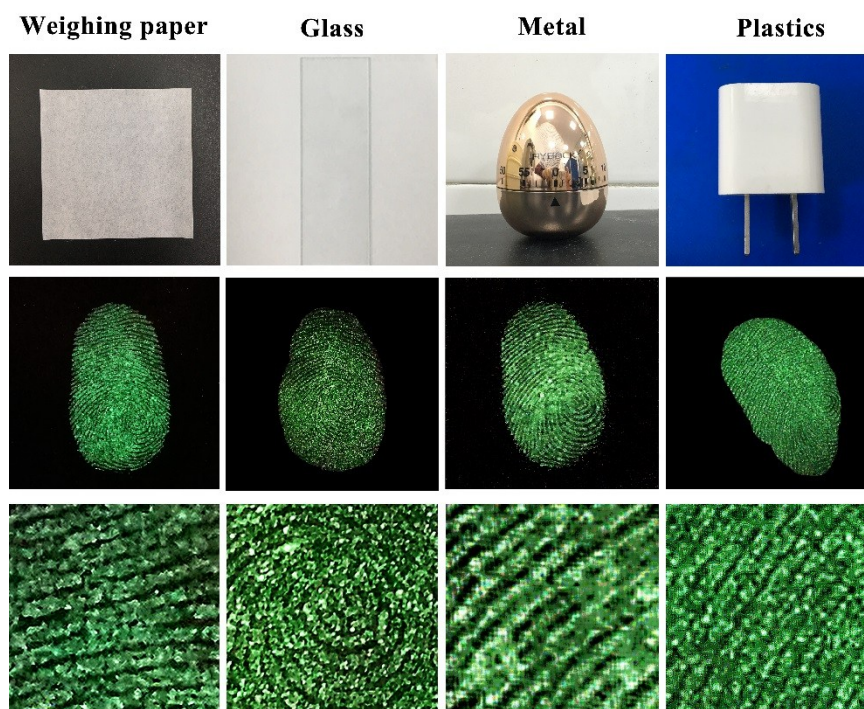
**Figure S9.** Corresponding photographs of CDs@SiO<sub>2</sub> powder in different solvents under sunlight, excited with 365 nm UV lamp, and after switching off UV lamp.



**Figure S10.** (a) phosphorescence emission spectra of CDs@SiO<sub>2</sub> composite using analogues ETA as starting material. (b) phosphorescence emission spectra of CDs@SiO<sub>2</sub> composite using analogues DETA as starting material.



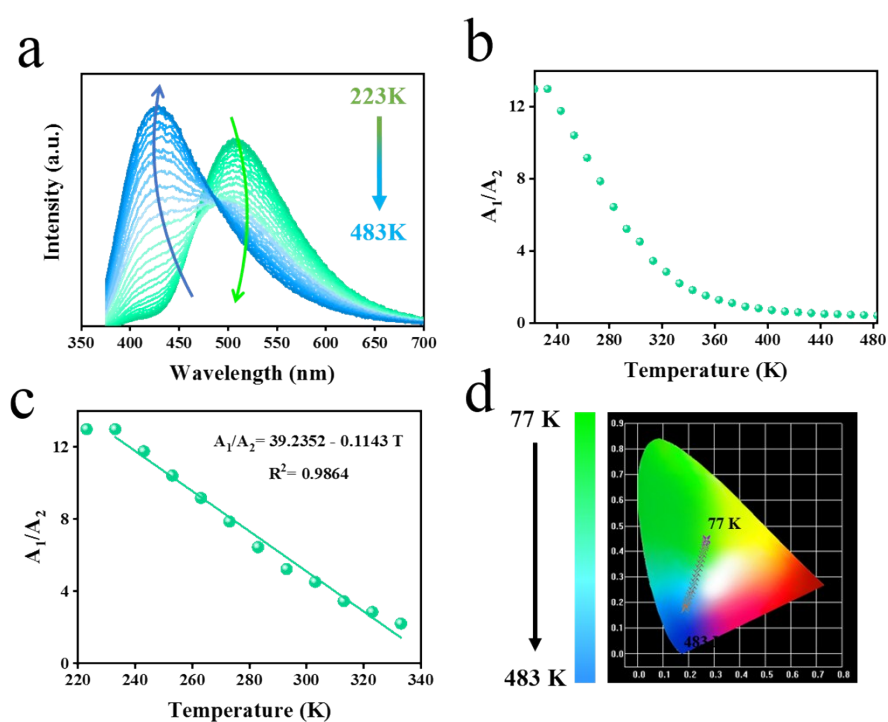
**Figure S11.** (a) phosphorescence emission spectra of CDs@SiO<sub>2</sub> composite using analogues EG as starting material. (b) phosphorescence emission spectra of CDs@SiO<sub>2</sub> composite using analogues CA as starting material



**Figure S12.** Photographs of CDs@SiO<sub>2</sub> powder stained fingerprints on different substrate with a mobile

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phone as the signal collector.



**Figure S13.** (a) Temperature-dependent afterglow emission spectra from 223-483 K recorded every 10 K under the excitation of 365 nm. (b) Temperature-dependent RTP/TADF emission ratio from 223 K to 483 K. (c) Temperature-dependent RTP/TADF emission ratio from 233 K to 333 K. (d) CIE coordinates of the afterglow emission of CDs@SiO<sub>2</sub> at the temperature range from 77 K to 483 K.

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**Table S1.** Specific surface area of the CDs@SiO<sub>2</sub> composite annealed at different temperatures.

Temperature (°C)	Specific surface area (m <sup>2</sup> /g)
400	155.66
500	162.38
550	180.78
600	122.87
700	56.33

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### Reference

- [1] G. Hu, Y. Sun, S. Wu, W. Li, C. Hu, J. Zhuang, X. Zhang, B. Lei, Y. Liu, *Nanotechnology*, **2019**, 30, 155601.