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Kilogram-scale synthesis of carbon dots with high-efficiency full-color solid-state fluorescence using an aggregation-induced emission strategy

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ABSTRACT

Carbon dots (CDs), one of the most promising luminescent nanomaterials, have attracted widespread concern due to their unique optoelectronic properties and widespread application. However, mass production of solidstate CDs with highly efficient full-color emission has rarely been reported, severely limiting their practical application in solid-state lighting and displays. Here, using an aggregation-induced emission (AIE) strategy, fullcolor solid-state fluorescent (SSF) CDs with quantum yields (QYs) of up to 55.3–27.9% have been prepared on a kilogram scale through one-step microwave-assisted pyrolysis of the same precursors. Detailed structure–property studies demonstrate that the SSF of CDs is derived from the suppression of surface carboxyl group motions and the decrease of the non-radiative rate in their aggregated state, and that their tunable emission is attributed to the excessive fluorescence resonance energy transfer (FRET) induced by the increasing number of aromatic rings on CD surfaces. Therefore, these CDs exhibit both the AIE enhancement and red shift throughout the entire visible spectrum in the solid state. Finally, because of their excellent AIE properties, these highly efficient SSF CDs have been applied to pattern encryption, large-area luminescent fibrous membranes (LFMs), light-emitting diodes (LEDs), and fingerprint detection.

1. Introduction

Over the past decade, carbon dots (CDs) have been most actively studied in imaging, sensing, light-emitting devices, and security systems owing to their outstanding merits of facile preparation, fascinating optical properties, excellent biocompatibility, and low cost [1–4]. However, similar to organic fluorescent molecules, most CDs are quenched in the solid state due to the direct π - π stacking interactions or excessive fluorescence resonance energy transfer (FRET) between them, thus limiting their direct application in lighting and display devices [5–8]. To overcome this severe aggregation-caused quenching (ACQ) effect, previous researchers have tried to disperse CDs into suitable solid matrices (polymers, inorganic salts, silica, *etc.*), or to modify their surfaces with functional ligands (alkyl amines, chlorosalicylaldehyde, dithiosalicylic acid, *etc.*) [9–13]. The former strategy produces very large particles or even bulk materials with insufficient photoluminescence (PL) output because the ACQ effect is still present [14–16]. The latter requires

complicated procedures such as time-consuming reactions, cumbersome post-treatment, and tedious purification processes (e.g. dialysis, and gel column chromatography), resulting in the production efficiency of CDs being extremely low (less than 1 gh⁻¹) [17–19]. Both the two approaches cannot produce a large number of CDs with highly efficient solid-state fluorescence (SSF) in a short time. In addition, the PL emission mechanisms for the SSF properties of CDs are currently unclear [20]. Thus, the mass production of CDs with full-color SSF is difficult to realize by predictable manipulation of experimental conditions.

The aggregation-induced emission (AIE) effect, as the opposite phenomenon of ACQ, has been used as an efficient strategy to prepare CDs with SSF [21]. The resulting AIE CDs are self-quenching resistant in the solid state without any post-treatment and can be quickly precipitated as powder by adding a poor solvent. These unique advantages not only maximize the SSF of CDs but also increase their product yield, giving the AIE approach enormous potential in mass-production of CDs with highly efficient SSF in a single step. However, such a strategy has

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Received 20 July 2023; Received in revised form 13 September 2023; Accepted 1 October 2023 Available online 11 October 2023 1385-8947/© 2023 Elsevier B.V. All rights reserved. not been implemented for the mass production of CDs yet [22]. What's more, most AIE CDs emit solely blue or yellow fluorescence, and it is difficult to tune their emission wavelengths. By far, only a few research groups have succeeded in the preparation of AIE CDs with tunable multicolor emissions by changing carbon sources [23]. In 2021, Hu's group synthesized green-to-red emitting AIE CDs through solvothermal treatment of dithiosalicylic acid and amines with different numbers of nitrogen atoms [21]. Most recently, Lu's group prepared a series of fullcolor emitting AIE CDs by modifying previously prepared CDs with different salicylaldehyde-type ligands [5]. Despite these efforts, the final AIE CDs still can't overcome the above technical obstacles because of the following reasons. (1) these CDs are not panchromatic or require timingconsuming post-treatment (more than 20 days); (2) the highest quantum yield (QY) of these CDs powder is only 11.7 % in the state-of-the-art; (3) different carbon sources were used to change PL emission, which is not beneficial for analyzing the luminescence mechanisms [24]. Therefore, it is a real challenge to develop a simple one-step preparation of AIE CDs with highly efficient full-color SSF from the same precursors, which is also important for the large-scale production and practical applications of SSF CDs, as well as for understanding their emission mechanism intrinsically.

In this work, we report one-step synthesis of AIE CDs with full-color SSF properties via microwave-assisted pyrolysis of 1,2,4,5-Benzenetetracarboxylic anhydride (PMDA) and o-phenylenediamine (o-PDA) for the first time. Taking advantage of their AIE properties and high-efficiency energy output of microwave irradiation, these SSF CDs can be obtained in kilogram quantities at ambient pressure after the amplification of the reactants, which is a breakthrough in the large-scale production of fluorescent CDs. The detailed investigation verifies that, in the aggregated state, the PL redshift is derived from the FRET induced by the increasing number of aromatic rings on CD surfaces, while the AIE phenomenon is attributed to the suppression of surface carboxyl group motions and the decrease of the non-radiative rate, both of which synergistically contribute to the highly efficient SSF (55.3-27.9 %) of CDs over the entire visible spectrum. Based on these findings, a reasonable PL mechanism was proposed. Due to their AIE properties, these CDs are used in pattern encryption, luminescent fibrous membranes (LFMs), light-emitting diodes (LEDs), and fingerprint detection, demonstrating their immense application potential in various fields.

2. Experimental section

2.1. Materials

1,2,4,5-Benzenetetracarboxylic anhydride (PMDA), o-phenylenediamine (o-PDA), polyvinyl alcohol (PVA), ethanol, dimethyl sulfoxide (DMSO), and dioctyl sulfosuccinate sodium salt (AOT) were of analytical grade and purchased from Aladdin Chemicals. GaN LEDs chips without phosphor coating were acquired from Advanced Optoeletronic Technology CO., Ltd. All chemical reagents were used as received without any purification. Ultrapure (Milli-Q) water was used throughout the study.

2.2. Synthesis of AIE-CDs with full-color SSF

These AIE-CDs with tunable SSF from blue to red were prepared from PMDA and o-PDA through a one-step microwave-assisted method. In a typical process, 4.81 mmol of PMDA and 0.46 mmol of o-PDA were dissolved in 15 mL of ethanol aqueous solution (volume ratio of ethanol to water is 20:1) in a 25 mL beaker with stirring for 10 min. The resulting solution was then heated in a domestic microwave oven (MKX-H2G1A) at 750 W for 10 min. After the reaction, the solution volatilized naturally and 10 mL of DMSO was added to form a CD solution, which was centrifuged at 3000 rpm for 10 min to remove large particles. Finally, 100 mL of water was added to the supernatant and the precipitated powders were filtered and dried to obtain B-CDs. Other samples were prepared using the same procedures as above except for different o-PDA amounts and reaction times. Specifically, 0.56 mmol of o-PDA and 12 min reaction time were used for G-CDs. 0.62 mmol of o-PDA and 12 min reaction time were used for GY-CDs. 0.91 mmol of o-PDA and 16 min reaction time were used for Y-CDs. 1.41 mmol of o-PDA and 20 min reaction time were used for O-CDs. 4.18 mmol of o-PDA and 20 min reaction time were used for R-CDs.

2.3. Kilogram-synthesis of AIE-CDs with full-color SSF

5.94 mol of PMDA and 0.57 mol of o-PDA were dissolved in 3000 mL of ethanol aqueous solution (volume ratio of ethanol to water is 20:1) in a 5000 mL beaker with stirring for 10 min. The resulting solution was then heated at 750 W for 10 min in a domestic microwave oven (MKX-H2G1A). After the reaction, the solution volatilized naturally and 1000 mL of DMSO was added to form a CD solution, which was centrifuged at 3000 rpm for 10 min to remove large particles. Finally, 9000 mL of water was added to obtain B-CDs on a kilogram scale. For G-, Y-, and R-CDs, the preparation procedures remained unchanged except that the o-PDA amounts were increased according to their respective ratios described in Figure S1.

2.4. Preparation of the security ink

The DMSO solutions respectively containing B-, G-, Y-, and R-CDs (0.2 mg/mL) can be directly used as security inks for anti-counterfeiting.

2.5. PVA/CDs fibrous membranes prepared by electrospinning

15 wt% PVA solution was prepared by dispersing 1.5 g of PVA (Mn = 72600–81400) and 0.04 g of dioctyl sulfosuccinate sodium salt (AOT) powder into 8.5 g of DMSO with vigorous stirring at 95 °C for 2 h. Then, 0.05 g of B-CDs, 0.10 g of G-CDs, 0.34 g of Y-CDs, and 0.40 g of R-CDs powder, respectively, were added to the PVA solution under vigorous stirring. The resulting homogenous solution was used for electrospinning. The distance between the burette tip and receiver was fixed at 15 cm and the high-voltage supply was fixed at 30 kV.

2.6. Fabrication of multicolor and white LEDs

Commercially available UV chips emitting 380 nm light with an operating voltage of 3.0 V were used as an excitation light source. For B-, G-, Y-, and R-CDs-based LEDs, 0.05 g of corresponding CDs powders were evenly mixed in 0.1 g of epoxy glue (volume ratio of A and B is 1:4). The CDs-epoxy glue mixtures were then dropped onto the LED chip and cured in an oven for 4 h at 60°C. Finally, the multicolor LEDs were obtained. To fabricate WLEDs, different weight ratios of B-CDs, G-CDs, and R-CDs were dispersed in epoxy glue following similar procedures for the fabrication of multicolor LEDs.

2.7. Latent fingerprint imaging

The latent fingerprint (LFP) samples were collected from the same donor, and prepared by lightly rubbing the finger on the forehead before touching some typical substrates. The R-CDs powders were then scattered uniformly onto the LFPs by the classical powder-dusting method, and the excess powders were removed with a feather brush. Finally, the fingerprint images were captured with a digital camera under irradiation of UV light (365 nm).

2.8. Characterization techniques

Transmission electron microscopy (TEM) and high-resolution TEM (HRTEM) images of CDs were observed on an FEI-Tecnai G2 apparatus at an acceleration voltage of 200 kV. Ultraviolet – visible (UV – vis)



Fig. 1. (a) Schematic illustration of the synthesis of SSF CDs. (b) Photographs of polychromatic SSF CDs under daylight (upper) and UV light (bottom), respectively. (c, d) Photographs of the four selected SSF CDs under UV light and visible light taken from the side (top) and top (lower) angles, respectively. (e) The normalized absorption spectra, (f) PL excitation spectra, (g) PL emission spectra, and (h) time-resolved PL spectra of the four selected CDs powders. (i-l) PL emission spectra of the selected B-CDs, G-CDs, Y-CDs, and R-CDs powders, respectively.

absorption spectra were monitored by a Shimadzu UV-3101 PC spectrophotometer. PL spectra of CDs were taken using an F-4600 Hitachi fluorescence spectrometer. Fourier-transform infrared (FTIR) spectra were obtained on a Magna IR-560 FTIR spectrometer. Raman spectra were measured with an XploRA Raman spectrometer under 785 nm of laser irradiation. X-ray photoelectron spectra were detected with an AXIS Ultra DLD spectrometer. Time-resolved fluorospectroscopy was performed using an FLS 920 spectrometer. The NMR spectra were carried out with a Bruker Avance III (600 MHz) spectrometer and DMSO-d₆ was used as the solvent. The CDs-based fibrous membranes were prepared using an electrospinning apparatus (TL-Pro-BM). The emission spectra, Commission Internationale de L'Eclairage (CIE) coordinates, color rendering index (CRI), and correlated color temperature (CCT) of the LEDs were obtained on a HAAS-2000 spectroradiometer.

3. Results and discussion

The CDs were synthesized by a facile one-step microwave-assisted

heating of an ethanol aqueous solution of PMDA and o-PDA under ambient conditions, followed by precipitation with water and separation by centrifugation (Fig. 1a). The key step in the preparation of full-color emitting CDs was the modulation of functional groups on CD surfaces, which was achieved by adjusting both the molar ratio of PMDA to o-PDA and the reaction time, as shown in Fig. S1. After optimizing the reaction conditions, six kinds of CDs powders with blue, green, green-yellow, yellow, orange, and red luminescence were obtained, which were denoted as B-CDs, G- CDs, GY-CDs, Y-CDs, O-CDs, and R-CDs, respectively. These CDs samples visually exhibited gradually deepening colors from light yellow to brown in visible light, and tunable PL emissions across the whole visible range under a 365 nm of ultraviolet (UV) lamp (Fig. 1b and Figs. S2-S3). Excitingly, this synthetic method enables the kilogram-scale synthesis of CDs after amplifying the reactants due to its fast reaction, easy purification, efficient separation, and normal pressure. Taking B-, G-, Y-, and R-CDs as examples, the yield of the four typical CDs could reach about 0.97 kg in 1 h (Fig. 1c-1d and Fig. S4), which benefits further mechanistic investigations and large-scale



Fig. 2. (a, b) Images of the four selected CDs dissolved in DMSO and DMSO/water (10 %/90 %) mixed solution under visible and UV light, respectively. (c-f) The normalized PL spectra of the selected CDs dissolved in water/DMSO mixed solutions with different water contents. (g) Images of the four selected CDs dissolved in DMSO with different concentrations under UV light. (h-k) The normalized PL spectra of the selected CDs dissolved in DMSO at different concentrations. (l) Overlap between the PL spectra of the four diluted CDs solutions (0.05 mg/mL) and the absorption of their concentrated solutions (40 mg/mL).

applications of SSF CDs in various fields.

The optical properties of the four CDs powders were investigated carefully. In their UV-vis absorption spectra shown in Fig. 1e, the four samples show distinct absorption maxima/bands centered at 344, 376, 412, and 465 nm, respectively, which are often ascribed to the $n-\pi^*$ transitions of the surface states with many C=O, C-O, and C-N structures [25-27]. In Fig. 1i-1 l, the PL spectra exhibit the maximum emission wavelengths of the four CDs located at 443, 509, 563, and 613 nm for B-CDs, G-CDs, Y-CDs, and R-CDs, respectively. Meanwhile, it is not difficult to observe that, from B-CDs to R-CDs, the excitation dependence of PL changes from dependent to independent and the narrowed full width at half maximum (FWHM) increases from 75 to 101 nm, both of which can be attributed to their increasing surface functionalized passivation [28–30]. The excitation spectra of the four samples in Fig. 1f are found to overlap well with their corresponding low-energy absorption bands, contributing to the blue, green, yellow, and red light emissions. By using an integrating sphere under their optimal excitation wavelengths, the absolute PL quantum yields (QYs) of the four CDs were measured to be 55.3 % for B-CDs, 48.8 % for G-CDs, 36.7 % for Y-CDs, and 27.9 % for R-CDs, which are the highest values for CDs with full-color SSF [31,32]. Furthermore, the PL decay curve of each sample is fitted by a singleexponential function (Fig. 1h), which gives average lifetimes of 5.16, 2.6, 2.28, and 1.89 ns for B-CDs, G-CDs, Y-CDs, and R-CDs, respectively, implying that all of the CDs underwent the single luminescence processes [33]. The above data clearly demonstrate that the four selected CDs possess uniform and intense SSF properties and that their tunable full-color emissions result from the change of surface states regulated by both the different molar ratios of o-PDA in the reactants and the reaction time.

To disclose their solid-state PL origins, the optical properties of the four CDs dissolved in dimethylsulfoxide (DMSO) solutions were investigated in detail. In Fig. S5, the UV-Vis absorption spectra show that these dilute CD solutions have enhanced and red-shifted absorption in the visible region from B-CDs to R-CDs. Interestingly, the normalized PL spectra in Fig. S6 display that these CD solutions are blue emissions with an increased FWHM with their PL redshift. These spectral changes again prove that our samples have increasing surface state transitions from B-CDs to R-CDs [34], similar to their solid-state counterparts. Next, the PL QYs and PL lifetimes of the four diluted CDs solutions were measured. The obtained PL QYs are in the range of 13.9-17.2 %, which is much lower than those in the solid state, indicating that the four CDs have AIE effects [35-37]. The PL lifetimes of the four dilute CD solutions are measured to be between 4.78 and 12.79 ns using a monoexponential fit. According to the relevant equations [21], the K_r (radiative rate constants) and K_{nr} (non-radiative rate constants) of the four CDs in solution and solid are then calculated and listed in Tab. S1. It can be seen that the K_r values in the solid state are much higher than in the solution state, which conforms to the basic rules of AIE activities [38,39]. Moreover, the viscosity- and temperature-dependent emission spectra of the four CD solutions were collected. The measurements in Figs. S7 and S8 show



Fig. 3. (a-d) TEM and HRTEM images (inset) of the four selected B-CDs, G-CDs, Y-CDs, and R-CDs, respectively. (e-h) AFM images of the four selected B-CDs, G-CDs, Y-CDs, and R-CDs, respectively. Black lines are the height-profiles analysis along the corresponding lines in Fig. 2e–h. (i) Raman spectra, (j) FTIR spectra, and (k) 13C NMR spectra of the four selected CDs.

that the PL intensity of these CDs increases significantly with the addition of the PVA and cooling of the samples, indicating that the restriction of the intramolecular motion (RIM) process is responsible for the AIE effect [35]. Therefore, it is demonstrated that the SSF of these CDs is due to the AIE effect rather than the intra- and inter-molecular hydrogen bond and crystallization because the R-CDs have almost no PL changes after alkali treatment and grinding (Fig. S9).

The AIE behavior of these CDs was further investigated by recording their optical properties in water/DMSO mixed solutions with different water contents. As shown in Fig. S10, when the water volume is less than 30 %, these CD solutions appear transparent under daylight and glow blue under UV light at the same concentration of 0.2 mg/mL. As the water volume is gradually increased, these homogeneous solutions are transformed into turbid solutions with suspension, implying that the water as a poor solvent could cause the aggregation of CDs with random orientation [40], which is verified by TEM images (Fig. S11). Meanwhile, the absorption and emission of these CDs solutions are accompanied by different degrees of enhancement and redshift with increasing water volume (Figs. S12-S14), suggesting that the widely recognized and accepted AIE enhancement and redshift coexist in our samples. In the 90 % of water volume ratio, these aggregated CDs nanoparticles could precipitate as powders at the bottom of solutions after standing for 20 min and emit intense blue, green, yellow, and red PL for B-, G-, Y-, and R-CDs, respectively (Fig. 2b). The time-resolved PL spectra of these CDs solutions with different water fractions show that their average lifetimes decrease with increasing water fractions (Fig. S15), confirming

the formation of alternative recombination channels in the aggregated state [41]. These phenomena confirm that CDs aggregation not only restricts the motions of surface functional groups but also alters their optical band gaps, so as to achieve the high-efficiency SSF emission of CDs over the entire visible range.

Subsequently, the AIE redshift mechanism of these CDs was deeply elucidated by studying the effect of CD concentration on their optical properties. From Fig. 2g and Fig. S16, it is clear that the four CD solutions exhibit concentration-dependent absorption and emission. Specifically, as the CD concentration gradually increases, the absorption spectra of the concentrated B-, G-, Y-, and R-CD solutions (Fig. S17) are significantly broadened and extend into longer wavelengths compared to those of their dilute solutions. The maximum PL emission peaks of the B-, G-, Y-, and R-CDs are redshifted by 12, 63, 116, and 152 nm, respectively (Fig. 2h-2 k and Fig. S18). Moreover, the time-resolved PL spectra of the four CDs show that their average lifetimes are much higher in solution than in solid (Fig. S19), suggesting the occurrence of FRET with increasing aggregation degree [42]. To determine their optical band gaps (Eg), the absorption spectra of the four CDs were further converted into Tauc plots of $(\alpha h\nu)^2$ versus $h\nu$, where α , h, and ν represent the absorption coefficient, Planck constant, and light frequency, respectively. In the Tauc plots (Fig. S20), all the E_g values, which are the intercepts on the x-axis, decrease with increasing CD concentration. At a concentration of 40 mg/mL, the Eg values of the four CDs were narrowed by 0.76, 1.27, 1.31, and 1.63 eV for B-, G-, Y-, and R-CDs, respectively, reflecting that the effect of increasing concentration on the optical band



Fig. 4. (a-d) High-resolution XPS C 1 s, N 1 s, and O 1 s spectra of the B-CDs, G-CDs, Y-CDs, and R-CDs, respectively.

gaps of these CDs increases from B-CDs to R-CDs (**Tab. S2**). These results clearly demonstrate that the concentration-dependent redshift is due to the FRET phenomenon [13]. In addition, the FRET efficiency of these CDs was assessed by calculating the spectral overlap between the absorption of concentrated solutions (40 mg/mL) and the emission of dilute solutions (0.05 mg/mL). The calculated overlap is 8.41 %, 40.23 %, 68.88 %, and 90.44 % for B-, G-, Y-, and R-CDs, respectively (Fig. 21). Among them, only the overlap of the B-CDs is lower than 30 %, which does not satisfy the condition of FRET [43], resulting in the B-CDs remaining unchanged in PL color even if they suffer from severe aggregation. As a result, the observed tunable SSF colors from blue to red are ascribed to the different FRET efficiency of these CDs.

To further confirm the relationship between the structural composition and fluorescence mechanism of these CDs, their sizes and morphologies were analyzed in detail by transmission electron microscopy (TEM), atomic force microscopy (AFM), and scanning electron microscope (SEM). As shown in Fig. 3a-3d, the TEM images reveal that these AIE CDs are homogeneous and well-dispersed nanodots, with similar average sizes of about 2.5 nm (Fig. S21). The high-resolution TEM (HR-TEM) pictures provided in the insets show that all the CDs have only one type of well-resolved lattice fringe with an in-plane lattice spacing of 0.21 nm, corresponding to (100) planes of graphite [44]. In Fig. 3e-3 h, the AFM images of the four CDs estimate their average topographic heights to be about 3.0-3.5 nm, which is larger than the diameters from TEM, suggesting the presence of polymer chains shells on CD surfaces [15]. The SEM images in Fig. S22 illustrate that all four CDs are aggregated quasi-spherical particles in the solid state with no obvious change in particle size and morphology. These data clearly indicate that the PL redshift from B-CDs to R-CDs is not due to quantum size effects [45]. The X-ray diffraction (XRD) patterns in Figs. S23 and S24 for these CDs powder exhibit that the diffraction peak at around 25° gradually decreases from B-CDs to R-CDs, accompanied by the appearance of additional sharp peaks, indicating that new crystalline domains are created by the formed polycyclic aromatic groups on the CD surfaces [5,25,46]. The Raman spectra of the four samples in Fig. 3i display two typical peaks at 1388 and 1593 cm^{-1} , which are ascribed to



Fig. 5. Schematic mechanism of these SSF CDs with aggregation-induced full-color emission.

the disordered sp³ carbon (D band) and graphitic carbon network (G band), respectively [2,47]. The intensity ratio of I_D/I_G increases from 1.38 of the B-CDs to 2.92 of the R-CDs (**Tab. S3**), which means that the observed PL redshift is closely related to the amorphous structures of CDs, in agreement with the TEM, AFM, and XRD results.

The surface functional groups and chemical compositions of the four CDs are further characterized by Fourier transform infrared (FTIR) spectra, X-ray photoelectron spectroscopy (XPS), and nuclear magnetic resonance (NMR). In the FTIR spectra (Fig. 3j and Fig. S25), the four samples show intense vibrational absorption of O-H/N-H at 3250-3570 cm⁻¹, –COOH at 1776 cm⁻¹, C=N at 1689 cm⁻¹, C-N at 1477 cm⁻¹, and C–O at 1124 cm⁻¹ respectively [30], confirming the presence of many polar functional groups on CD surfaces. Remarkably, both the typical vibrational absorption of C=C and C=N gradually increase as the stretching vibration intensity of -COOH decreases significantly from B-CDs to R-CDs, indicating that an increasing number of N-containing polyaromatic groups are attached to the CD cores through the dehydration and carbonization reactions [33]. The full XPS spectra (Fig. S26) reveal three peaks at 284.7, 400.5, and 531.8 eV, demonstrating that the elements of C, N, and O all exist in the four samples. The element content of the four CDs increases from 0.61 to 0.76 for C but decreases from 0.33 to 0.12 for O in the order of their redshifted emission wavelengths. In the high-resolution spectrum (Fig. 4), the C1 s spectra are deconvoluted into four peaks at 284.4, 285.5, 286.6, and 288.2 eV, assigned to C-C/C=C, C-N, C-O, and C=O, respectively [48]. The N 1 s spectra are resolved into three peaks at 398.4, 400.2, and 401.4 eV, representing C-N=C, N-(C)₃, and N–H, respectively. The O 1 s spectra are divided into two peaks at 531.6 for C=O and 533.3 eV for C-O [49]. The corresponding data in **Tab. S3** illustrates that the proportion of C=C increases from 0.53 to 0.61 and that the proportion of C=N-C increases from 0.07 to 0.29, revealing the increase in the N-containing aromatic structure content on the surfaces of these CDs. Besides, NMR spectroscopy was also employed

to distinguish the surface functional groups of CDs. In the ¹³C NMR spectra (Fig. 3k), the four CDs show the signals at 30–45, 120–140, and 160–180 ppm, which belong to the C–C/C–N/C–O carbons, carboxyl carbons, and aromatic carbons, respectively. The ¹H NMR spectra in Fig. S27 contain the C–H protons in 2–3 ppm, the aromatic protons in 6–8 ppm, and the protons attached to carboxyl groups in 13–14 ppm [30]. Clearly, both the ¹³C NMR and ¹H NMR spectra display an increase in the relative intensity of the conjugated sp² structures to carboxyl groups from B-CDs to R-CDs, again proving that the number of aromatic structures on the CD surfaces increases with their distinct PL redshift. Based on the above characterizations and discussion, it is concluded that these CDs consist of large-sized graphitized carbon cores covered with abundant functional groups and that the increasing amount of aromatic structure groups on their surfaces results in the observed PL redshift from blue to red.

It has been reported that the functional groups attached to the sp²hybridized carbons could induce significant local distortions and then create numerous new sub-levels between the highest occupied molecular orbital and the lowest unoccupied molecular orbital energy levels in the original materials [50]. As a result, many types of radiative recombination could occur, leading to a remarkable redshift in absorption and emission derived from the surface state. In our case, we believe that the different surface states that are induced by the increased amount of o-PDA are responsible for the observed redshift in absorption and emission. Moreover, in the control experiments, the PMDA derived CDs are blue emissive in the solid state with a QY of 16.5 %, while no solid-state PL was observed in CDs derived from o-PDA (Fig. S28). Then, the optical properties of CDs dispersed in DMSO/water mixed solvents were investigated. In Figs. S29 and S30, as the water content increases from 10 % to 90 %, the PMDA-derived CDs show an obvious enhancement in both absorption and emission intensity, but no PL peak redshift, indicating that their AIE activities are similar to those of the B-CDs. In



Fig. 6. (a) Images of letters written with different CDs-based inks under UV light before and after sparing water, respectively. (b) Optical and (c) fluorescence microscopy images of the as-prepared different CDs-based fibrous membranes under UV light. (d-f) The CIE color coordinates of the cool, standard, and warm WLEDs. The insets are images of the cool, standard, and warm WLEDs. (g) Images of the R-CDs powder-stained LFPs on different substrates under UV light. Images for (j1–6) are magnified images showing six kinds of details as marked in LFPs.

Fig. S31, the time-resolved PL spectra of the CDs are also fitted by a single exponential function and give an increased lifetime from 0.52 to 2.97 ns with increasing water content. These data suggesting that the PMDA is the critical component in the preparation of CD platforms with AIE characteristics and that o-PDA acts as a surface ligand to modify the formed AIE CD cores. Based on these reported conclusions and the above data analysis, a convincing PL emission mechanism can be proposed. As schemed in Fig. 5, these CDs consist of large-sized graphitized carbon cores coated with abundant functional groups. Since a large number of –COOH is retained and distributed on the CD surface, the $n - \pi^*$ transitions of C=O become dominant, so that these CDs emit blue PL in the dispersed state [29]. With the increase of aromatic groups in these CDs, numerous localized electronic states below the π^* state are expected to form, causing a remarkable redshift in their absorption and emission. Therefore, the CDs with an increased amount of aromatic structures are able to act as the FRET acceptors, contributing to the increased spectral overlap from B-CDs to R-CDs. When the distance between adjacent CD

nanoparticles was reduced to less than R₀ (FÖrster radius) by adding water or increasing the concentration, the FRET would take place with different efficiencies, resulting in tunable PL emission across the visible spectrum [13]. Meanwhile, the CD aggregation would hinder the rotation of their surface groups including the carboxyl and benzene ring, and weaken the non-radiative transition, thereby enhancing the SSF emission [19]. Both the AIE and FRET effects synergistically produce high-efficiency SSF emission of CDs from blue to red. To further confirm such a conclusion, the tuning experiment was carried out under the same conditions as for the R-CDs except that the amount of o-PDA was increased to 4.81 mmol. The resulting CDs are emissive in the solid state with a PL peak at 639 nm, which is redshifted by 26 nm compared to the R-CDs (Fig. S32). Meanwhile, their solid state QY decreases from 27.9 % to 3.6 % due to the FRET effect. These data are consistent with the conclusion regarding the AIE mechanism of these SSF CDs.

Considering their aggregation-induced full-color emission, high QY, and excellent photostability (Figs. S33 and S34), these CD powders

were made for various applications. Firstly, utilizing their aggregationinduced multicolor emission in different solvent environments, four kinds of security inks were prepared by dissolving the B-, G-, Y-, and R-CDs in DMSO, respectively, which are used in order to write the letters "CUMT" on the filter paper. Under UV irradiation, these letters show the same blue color but present different luminous colors after water is introduced (Fig. 6a). Then, the multicolor returned to blue emission after the DMSO was sprayed again, thus realizing the higher levels of information encryption compared to CDs with single emission [51,52]. Secondly, due to their mass production and tunable SSF, these CDs were employed as phosphors to fabricate luminescent fibrous membranes (LFMs) and light-emitting diodes (LEDs). In Fig. 6b and 6c, large-area LFMs (30 cm \times 20 cm) with full-color emission were produced via single-nozzle electrospinning within 1 h, demonstrating a great potential to be used in wearable optoelectronics with less toxic than perovskite or Cd/Pb based quantum dots (QDs) [53]. In Fig. S35, the four types of monochrome LEDs based on the B-, G-, Y-, and R-CDs produce blue, green, yellow, and red light with peaks at 441, 508, 560, to 614 nm, respectively (Fig. S36). Their tunable emission also inspires us to construct all types of WLEDs with the B-, G-, and R-CDs in different mass ratios. When the mass ratio of B-, G-, and R-CDs is 1.4:1.0:0.7, a cool WLED is achieved with CCT of 9084 K, CIE coordinates of (0.28, 0.29), and CRI of 91 (Fig. 6d). When the mass ratio is fixed at 0.8:1.1:1.0, a standard WLED is obtained (Fig. 6e), which generates white light akin to natural sunlight [26]. The CCT (5227 K), CIE coordinates (0.32, 0.33), and CRI (92) of the pure WLED indicate its better performance than those of most CD-based WLEDs [54]. By changing the mass ratio of B-, G-, and R-CDs to 0.9:1.0:1.3, a warm WLED is obtained that emits comfortable and warm white light with CCT, CIE coordinates, and CRI of 3813 K, (0.40, 0.39), and 93, respectively (Fig. 6f). The three types of WLEDs exhibit tunable light emission spectra from blue to deep red (Fig. S37), high CCT comparable to semiconductor QDs [55], and remain almost unchanged in the emission intensity after continuous operation over 24 h (Fig. S38), demonstrating their excellent photostability. Finally, for the highly emissive R-CDs, we attempted to detect and identify the latent fingerprints using the typical powder dusting method, as they are much safer, clearer and more compatible than those fluorescent organic dyes [55]. As shown in Fig. 6g, LFPs are successfully detected under UV irradiation on common substrates such as glass, wood, and aluminum alloys. The ridge patterns of all samples, including arches, loops, and whorls, which are classified as level one, are clearly visible due to the high contrast of the fluorescence signals [56]. After enlarging the fingerprint pattern, six feature points are clearly observed: bifurcation, termination, island, eye, core, crossover, and scar. Overall, these full-color emitting CDs have great practical applications in the field of solid-state luminescence.

4. Conclusion

In summary, we have reported the one-step synthesis of AIE-type CDs with highly efficient full-color SSF using carefully selected PMDA and o-PDA as raw materials. Both the AIE characteristics and the efficient microwave-assisted heating work together to greatly improve the production efficiency of SSF CDs compared with conventional methods, enabling their synthesis on a kilogram scale in a short time, which is a huge advance in the mass production of CDs. The detailed data analysis proved that the aggregation of CDs could lead to the suppression of surface carboxyl group motions and the reduction of the non-radiative rate, so as to obviously increase their PL efficiency in solid state. Meanwhile, the increasing amount of aromatic structures on the CD surfaces would induce enhanced FRET efficiency, resulting in aggregation-induced red-shifted emission from B-CDs to R-CDs. Furthermore, these CDs were successfully applied to pattern encryption, large-area LFMs, LEDs, and fingerprint identification to meet different application requirements. This work not only provides an AIE methodology for the scaled-up synthesis of SSF CDs but also stimulates their mechanistic study and practical application in the future.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.cej.2023.146405.

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