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Article

High production-yield solid-state carbon dots with tunable photoluminescence for white/multi-color light-emitting diodes

Haoqiang Song [a], Xuejian Liu [a], Boyang Wang [a], Zhiyong Tang [b,c], Siyu Lu [a,b,*]

[a] College of Chemistry, Zhengzhou University, Zhengzhou 450001, China
[b] Henan Institute of Advanced Technology, Zhengzhou University, Zhengzhou 450001, China
[c] CAS Key Laboratory of Nanosystem and Hierarchical Fabrication, CAS Center for Excellence in Nanoscience, National Center for Nanoscience and Technology, Beijing 100190, China

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ABSTRACT

Plastic waste is generally resistant to natural degradation and has become a major environmental pollution problem globally. The pollution of ecosystems seriously affects the health and survival of organisms, including humans. Much attention has been paid to finding suitable ways to convert plastic waste into high-value-added carbon materials. To this end, we report the high production yield (60%–85%) of carbon dots (CDs) for solid-state fluorescence (SSF) obtained by a one-step solvothermal method using waste expanded polystyrene as the precursor. The SSF mechanism of the CDs was also explored. Their emission wavelength, with a large full width at half maximum of 150–200 nm, exhibited tunable photoluminescence from white to yellow and orange. CDs powder was used to fabricate single-component white and multi-colour light-emitting diodes on UV chips. Overall, plastic waste was converted into tunable solid-state fluorescent CDs powder, which has promising applications in carbon-based lighting, by a simple solvothermal method that provides a viable method for recycling plastic waste.

1. Introduction

The harm to the environment caused by plastic waste has become a global problem and is receiving increased research attention [1–5]. Plastic waste poses a threat to terrestrial, marine, freshwater ecosystems and large numbers of organisms, including humans [6,7]. Disposable medical devices, fast food boxes, and plastic bottles are common plastic waste; they are mainly composed of polyethylene, polyvinyl chloride, polystyrene, and other organic polymers. Although convenient, these objects become plastic waste, which causes serious pollution problems because it is difficult to degrade naturally. The production of plastics is increasing by several million tons per year. Polystyrene is one of the five general-purpose plastics, and its annual output is in the hundreds of millions of kilograms. Expanded polystyrene is an inert, hydrocarbon thermoplastic that is extensively used in packaging and thermal insulation. Its low density allows it to escape into the environment easily [8]. The convenient solution of burning waste expanded polystyrene (WEPS) causes air pollution and emits carbon dioxide, which contributes to climate change. Hence, a simple, convenient method needs to be developed to increase the added value of these waste materials [9]. We fabricated carbon dots (CDs) from WEPS and observed fascinating tunable photoluminescence (PL) in the solid state [10–12].

CDs are a promising new carbon-based material owing to their outstanding properties, including biocompatibility, chemical stability, fabrication from a wide range of precursors, low toxicity, and low cost [13–15]. Potential uses of CDs include catalysis [16,17], bioimaging [18–20], optoelectronics [21,22], and energy conversion [23–26]. However, the development of CDs still faces several important challenges [27]. First, their emission wavelengths are mostly in the blue or green regions, but many applications require longer (e.g., orange or red) or wider (e.g., full spectral range coverage) wavelength emissions [28,29], such as single-component white light-emitting diodes (WLEDs) with low corresponding color temperature (CCT) and high color rendering index (CRI). Second, the hydrophilic functional groups commonly present on the surfaces of CDs often cause difficulties in keeping CDs powders dry in common conditions [30]. Moreover, similar to organic molecules, solid-state CDs usually undergo self-quenching resulting from direct π–π interactions or excessive resonance energy transfer (RET) [31]. Although silica [32] and polymers [33] have been reported as solid matrices capable of supporting the incorporation of CDs and enabling or adjusting their solid-state fluorescence (SSF), they cannot fundamentally prevent quenching by aggregation. Therefore, there is need to develop solid-state CDs with long wavelength emissions and resistance to self-quenching.

* Corresponding author.
E-mail address: sylu2013@zzu.edu.cn (S. Lu).

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Lighting consumes almost one-fifth of the world's electricity. Modern lighting is designed to save energy by having high luminous efficiency [34]. Much effort has been spent on developing WLEDs owing to their potential in energy saving; they are efficient light-emitting devices with long service life and have disrupted the traditional incandescent lamp industry. Compared with traditional phosphors using rare-earth elements [35] or heavy metals [36], the stable optical properties, low cost, and low toxicity of CDs makes them attractive as WLEDs phosphors. At present, WLEDs are produced using multiple phosphors of different colors or yellow phosphors combined with blue or UV chips [37–39]. The former involves complex production processes, whereas the latter leads to a low CRI and high CCT. In contrast, the wide PL characteristics of CDs can be used directly to prepare single-component WLEDs with high CRI and low CCT. However, achieving white emission requires the emission bandwidth of CDs to be increased and the self-quenching caused by aggregation to be overcome, which are major challenges.

This work reports the use of WEPS to synthesize high-production hydrophobic CDs, which overcome self-quenching and emit tunable SSF with a large full width at half maximum (FWHM, 150–200 nm) by adjusting the CDs' degree of oxidation. Through controlling the red-shift caused by an appropriate degree of aggregation, we achieved white-light and multi-color emissions in organic media using only solid-state CDs. Moreover, WLEDs containing CDs phosphors showed good stability. Producing solid-state fluorescent CDs from plastic waste increases the added value of plastic waste and opens a new area of research into plastic waste treatment.

2. Experimental

2.1. Materials

Waste expanded polystyrene foam (WEPS), dichloromethane (DCE), HNO_3, polydimethylsiloxane (PDMS). Except WEPS, all chemicals reagents were analytical grade and used as received without further purification.

2.2. Preparation of luminescent CDs

CDs were synthesized by solvothermal method. Typically, 0.1 g WEPS were dissolved in 10 mL DCE to form a transparent solution. Then 5–30 μL HNO_3 were dropped into as-prepared solution. The mixture was transferred to a poly (tetrafluoroethylene) (Teflon)-lined autoclave (25 mL) and heated at 200 °C for 5 h. After the reactor was cooled to ambient temperature automatically, the solution was still transparent and its color turned into pale-yellow to brownish-red under daylight. The obtained solution was heated to remove DCE to obtain CDs. According to the SSF of CDs powder, these CDs are named white CDs (W-CDs), yellow CDs (Y-CDs) and orange CDs (O-CDs).

For more experimental details, please refer to the Supplementary materials.

3. Results and discussion

The CDs were prepared by a one-step solvothermal method by treating WEPS in dichloromethane (DCE) with a micro volume of HNO_3 at 200 °C for 5 h (Fig. 1a). The DCE had two important functions in the reaction: to dissolve the WEPS and to form a network of three-dimensional cross-links by linking the benzene rings of polystyrene in the presence of HNO_3. As the volume of HNO_3 increased, the color of the CDs solution gradually deepened from pale yellow to red–brown, and the production-yield of CDs gradually increased (60%–85%).

The CDs solution exhibited excitation-dependent PL (Fig. S1a–c online), and the UV–vis absorption spectrum showed a strong absorption at about 260 nm, which was assigned to the π–π* transition of the aromatic C=C bond (Fig. S2a–c online). Evaporating the solvent left solid-state fluorescent CDs. Increasing the volume

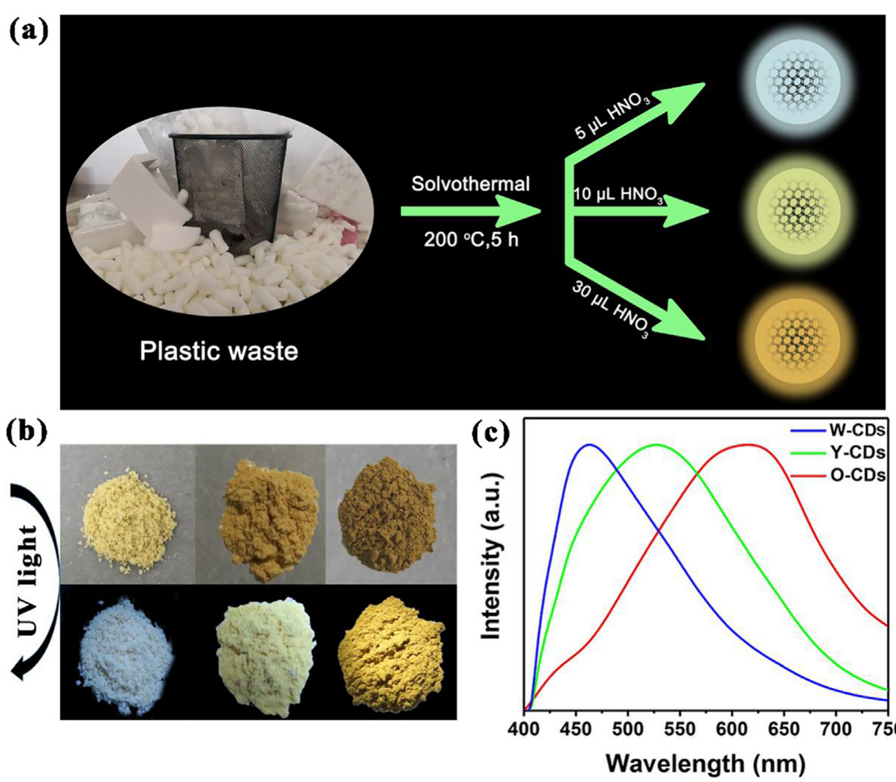

Fig. 1. (a) Schematic of the fabrication of solid-state fluorescent CDs. (b) Photographs of W-CDs, Y-CDs, and O-CDs under daylight (top) and 365 nm UV (bottom), and (c) their SSF spectra.

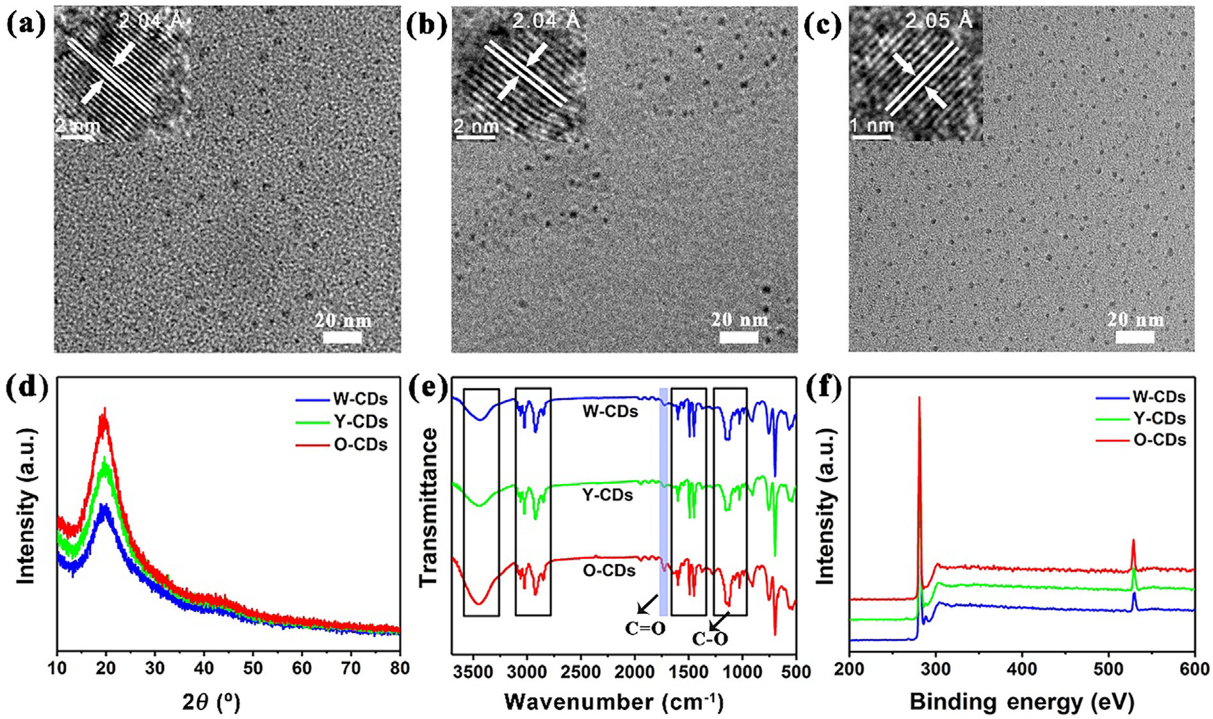

Fig. 2. TEM images of (a) W-CDs, (b) Y-CDs, and (c) O-CDs (insets: corresponding HRTEM images); (d) XRD patterns; (e) FT-IR spectra; and (f) full XPS spectra.

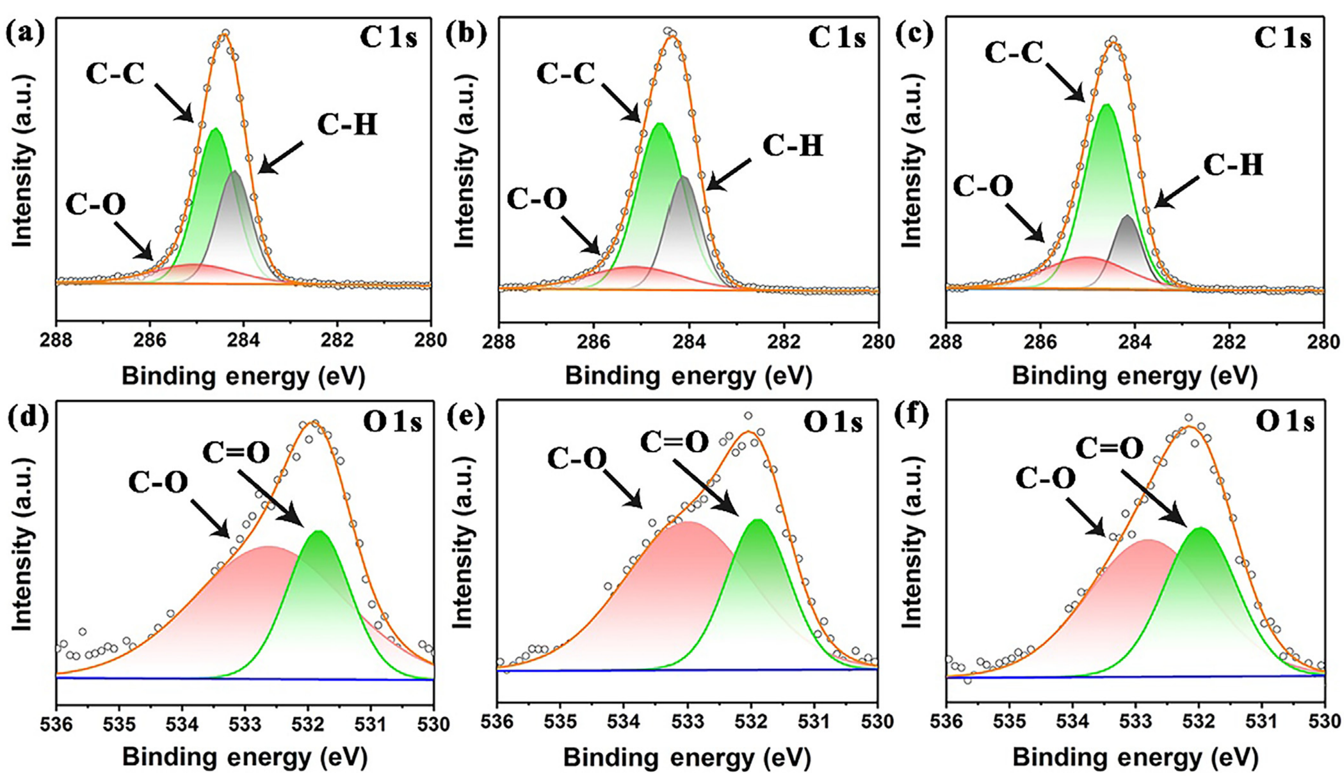

Fig. 3. XPS spectra. (a–c) C 1s and (d–f) O 1s spectra of W-CDs (left), Y-CDs (centre), and O-CDs (right).

of HNO_3 varied the CDs powder's emission from white, to yellow, and to orange under UV light (365 nm), and the powders were named W-CDs, Y-CDs, and O-CDs, accordingly (Fig. 1b). Their fluorescence emission centers were at 470, 530, and 630 nm, respectively (Fig. 1c). The FWHM of W-CDs was around 150 nm, whereas those of the other two powders were around 200 nm, almost covering the entire visible region (400–750 nm). The UV–vis absorption spectra of the CDs powder showed strong absorption in the UV region with partial absorption in almost the entire visible region (400–700 nm) (Fig. S3a–c online). The PL quantum yields (QYs) of the solid-state W-CDs, Y-CDs, and O-CDs under 365 nm excitation were measured to be 5.2%, 3.4%, and 3.1%, respectively.

Transmission electron microscopy (TEM) was used to characterize the microstructures of W-CDs, Y-CDs, and O-CDs (Fig. 2a–c). Each showed a uniform dispersion and spherical particles with approximate average diameters of 4.5, 3.5, and 2.3 nm, respectively. The size distributions are shown in Fig. S4a–c (online). High-resolution TEM (HRTEM) images showed that the three CDs had similar crystallinity with average lattice spacings of about 0.21 nm, corresponding to the (1 0 0) planes of graphite. A clear, strong, broad X-ray diffraction (XRD) peak centered at around 21° indicated that the CDs' had a graphitic structure (Fig. 2d), consistent with the HRTEM images. Fourier transform infrared (FT-IR) spectra characterized the surface states in the CDs (Fig. 2e). Two strong, broad absorption peaks at 1220–1020 and

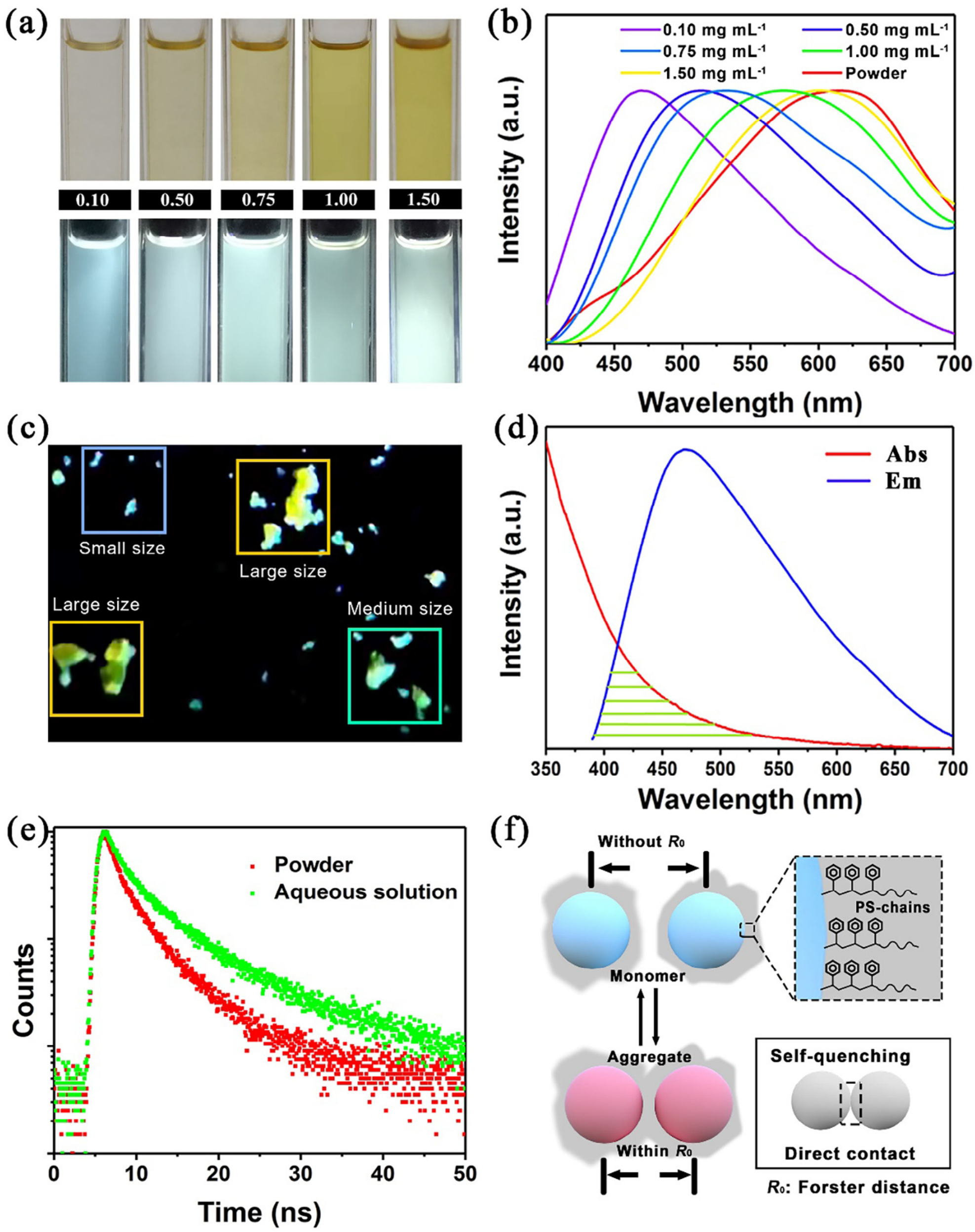

Fig. 4. (a) Photographs of O-CDs solutions of different concentrations under daylight (top) and UV light (365 nm, bottom). (b) Emission spectra of O-CDs in the solid-state and in various solutions (excited at 380 nm). (c) Fluorescence microscopy images of O-CDs powder in ethanol suspension. (d) Overlapping UV–vis absorption and emission spectra of a dilute solution of O-CDs (0.10 mg mL^{-1}). (e) Fluorescence lifetime spectra of O-CDs powder and dilute solution. (f) Schematic of CDs with self-quenching resistance and aggregation-induced red-shift. Inset: common self-quenching PL species.

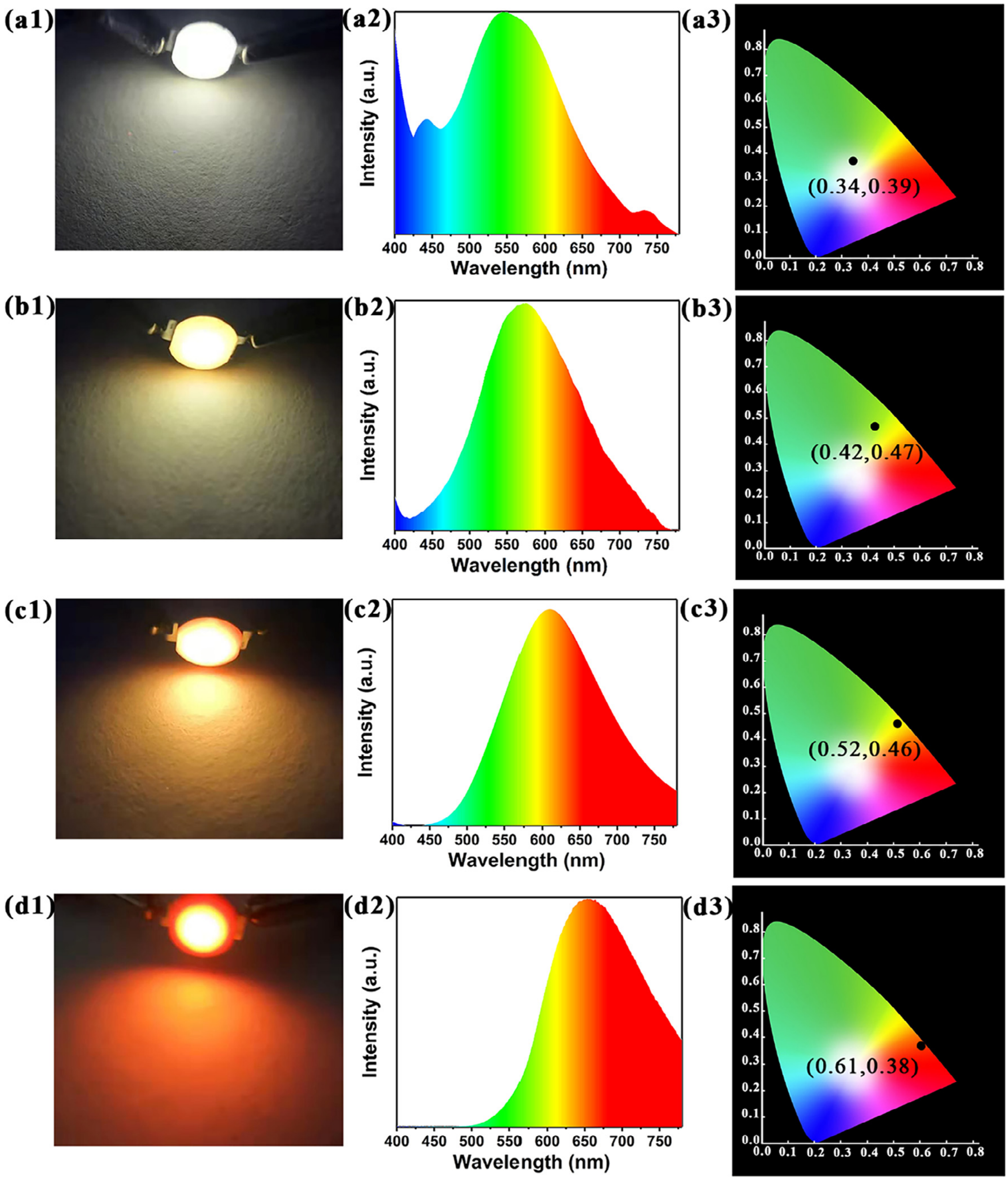

Fig. 5. PL photographs (left), emission spectra (centre), and CIE colour coordinates (right) of the (a1–a3) white, (b1–b3) warm white, (c1–c3) yellow, and (d1–d3) orange–red phosphor-based LEDs.

3600–3100 cm^{-1} are typical C–O and O–H stretching vibrations. The peaks at 1600, 1500, and 1450 cm^{-1} are C═C stretching vibrations; those at 696 and 756 cm^{-1} are due to hydrogen adjacent the benzene rings of polystyrene, implying that the CDs' surfaces allowed the attachment of polystyrene fragments [40]. The peak at 1730 cm^{-1} is from C═O stretching vibrations, and its gradual strengthening across the series of CDs reflects an increasing degree of oxidation accompanying the red-shift of the fluorescence. X-ray photoelectron spectroscopy (XPS) was used to characterize groups on the CDs' surfaces. The full spectrum clearly shows C 1s (284.6 eV) and O 1s (532.2 eV) peaks, suggesting that these were the CDs' main components (Fig. 2f). Analyzing the C and O contents (Table S1 online) showed that the O content of the CDs increased from 3.83% to 6.02% as the volume of HNO_3 increased, indicating the gradual increase in the degree of oxidation [41]. High resolution C 1s and O 1s spectra provided further insight into the changes in the degree of oxidation of the CDs. The C 1s spectra in Fig. 3a–c was deconvoluted into C–O (285.1 eV), C–C (284.6 eV), and C–H (284.1 eV) peaks [42]. The O 1s spectra contained C–O (532.8 eV) and C═O (531.9 eV) peaks (Fig. 3d–e). The gradually increasing intensities at 285.1 and 531.9 eV were due to the increasing degree of oxidation in the CDs (Tables S2 and S3 online), consistent with the FT-IR results. The increased degree of oxidation (i.e., more oxygen atoms contained in surface structures) may have reduced the band gap and caused the red-shift of the emission wavelength of the CDs [43,44].

In addition, the PL color in solution of O-CDs gradually changed from pale blue to blue–white and yellow–white as their concentration increased from 0.10 to 1.50 mg mL^{-1} (Fig. 4a), and the emission wavelength red-shifted greatly (from 470 to 630 nm) as the concentration increased from a dilute solution to the solid state (Fig. 4b). The fluorescence microscopy images in Fig. 4c of O-CDs powder in ethanol suspension show the change in fluorescence from pale blue to yellow-white as the aggregation size of the O-CDs increased, which was consistent with the above results, and confirmed that aggregation red-shifted the fluorescence [45]. Combining these observations and the spectral overlap between the absorption and emission spectra of the O-CDs (Fig. 4d), which can be expressed as the Förster distance (R_0) for convenience, suggests that a large red-shift caused by RET will occur when the small molecules are separated by less than R_0. Further confirmation of RET in O-CDs aggregates came from measuring the fluorescence lifetimes of the O-CDs powder and dilute solution (Fig. 4e). The longer average lifetime of the solution (5.32 ns) compared with the powder (3.63 ns) indicated RET, which probably competed with lifetime-shortening radiative transitions. The surfaces of the O-CDs retained polystyrene fragments, which may have controlled the interparticle distance by preventing direct contact, thereby preventing the O-CDs from fluorescence quenching and enabling them to achieve red-shifted emission through RET [46]. A possible mechanism of the resistance to fluorescence self-quenching and aggregation-induced red-shift is shown in Fig. 4f.

The unique optical properties of CDs make them useful in solid-state lighting. As the FWHM of the O-CDs was over 200 nm, covering almost the entire visible spectrum, they were fabricated into single-component WLEDs. Mixing O-CDs with polydimethylsiloxane (PDMS) solution produced a hybrid O-CDs/PDMS nanocomposite. A 365 nm light-emitting chip acted as an excitation source, onto which the nanocomposite was coated to form a device providing white illumination with CIE (Commission Internationale de l'Eclairage) coordinates of (0.34, 0.39), a CCT of 5199 K, and a CRI of 80.0 (Fig. 5a1–a3). A WLEDs prepared using O-CDs phosphors on a UV chip emitted close to pure white coordinates (0.33, 0.33), and maintained nearly 90% of its luminous intensity under continuous illumination for 10 h at 20 mA; thus, demonstrating good stability (Fig. S5 online). Furthermore, the luminous intensity varied almost linearly with current and Fig. S6 (online) shows the good current response. The concentration-dependent PL of the O-CDs solution suggests that the same effect could be achieved through controlling the quality ratio of O-CDs to PDMS. A series of O-CDs-based LEDs produced warm-white, yellow, and orange-red light with corresponding CIE coordinates of (0.42, 0.47), (0.52, 0.46), and (0.61, 0.38), respectively (Fig. 5b–d); their CCTs were 3660, 2385, and 1305 K, respectively. The other two CDs also formed multi-color LEDs when their concentration in PDMS was controlled (Table S4 online). Overall, these results show that white/multi-color LEDs can be prepared by controlling the ratio of CDs to PDMS. Therefore, plastic-based solid-state CDs represent significantly reduced production costs for phosphors.

4. Conclusions

In summary, we developed a simple, efficient one-step synthetic route to convert WEPS into solid-state fluorescent CDs with high production yield. By controlling the volume of HNO_3, solid-state CDs were produced with PL tunable from white to orange, and the FWHM of the emission wavelength varied from 150 to 200 nm. Powders of each CDs type could be applied individually to UV chips as the only phosphor to fabricate white, warm-white, yellow, and orange-red LEDs by adjusting the quality ratio of CDs to PDMS. The WLEDs showed good stability. Using WEPS derived CDs as phosphors in white/multi-color LEDs provides a promising approach to solving the problem of plastic waste.

Conflict of interest

The authors declare that they have no conflict of interest.

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Author contributions

Haoqiang Song and Xuejian Liu contributed equally. Haoqiang Song, Xuejian Liu and Boyang Wang contributed to the synthesis and characterization. Haoqiang Song and Xuejian Liu wrote the manuscript. Zhiyong Tang supported and supervised the research. Siyu Lu conceived the idea and organized the manuscript. All the authors contributed to discussion.

Appendix A. Supplementary materials

Supplementary materials to this article can be found online at https://doi.org/10.1016/j.scib.2019.10.006.

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Haoqiang Song is currently a M.S. student of College of Chemistry and Chemical Engineering, Henan Polytechnic University. He has been co-cultivating at Zhengzhou University since 2017 under the supervision of Prof. Siyu Lu and Baozhong Liu. His main research focuses on the synthesis of carbon-based materials and their electrocatalytic applications.

Siyu Lu is currently an associate professor of College of Chemistry and Molecular Engineering, Zhengzhou University. He received his Ph.D. degree in polymer physics and chemistry under the supervision of Prof. Bai Yang at Jilin University. His research interests relate to the design, preparation, properties of photoelectric nanocrystals (Carbon dots, Semiconductors, etc.) and their applications in catalysis, superconductivity, thermoelectricity, and structural prediction.