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# A review of fluorescent carbon dots: synthesis, photoluminescence mechanism, solid-state photoluminescence and applications in white light-emitting diodes

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**Abstract:** Carbon nanomaterials with a size of less than 10 nm, fluorescent carbon dots (CDs), have been extensively investigated, due to their excellent fluorescence tunability, good biocompatibility, wide range of precursors and low cost. Moreover, their simple preparation and excellent performance provide for a wide range of applications in the fields of optical sensing, energy storage, biomedical imaging, and white light-emitting diodes (WLEDs). A large number of solid-state photoluminescent CDs have recently been developed and used in WLEDs. The synthesis strategies of CDs are briefly summarized and their photoluminescence mechanisms are reviewed as well as the recent progress for their use in WLEDs. Finally, prospects for solving the current problems and challenges of CDs for WLEDs are briefly presented and discussed.

**Key words:** Carbon dots; Photoluminescence; Solid-state photoluminescence; WLEDs

## 1 Introduction

Working as a new type of high-efficient light-emitting source, white light-emitting diodes (WLEDs) open up a technical field for the lighting industry and are far superior to traditional incandescent lamps in terms of luminous efficiency and performance. Typically, there are two routes currently to fabricate WLEDs: Coating a variety of monochromatic phosphors on UV chips[1–2] or using phosphors by "blue light technology" to form white light[3]. Previously, rare earth phosphors were widely used as the mainstream luminescent material in WLEDs[4]. However, the high cost and toxicity of rare earth materials seriously hindere their further development. It is therefore of great significance to find a new green luminescent material with low cost, low toxicity, and with high photoluminescence (PL), so that it could be exploited on a large scale.

As a novel zero-dimensional (0D) carbon nanomaterials discovered in 2004[5], carbon dots consist of a carbonized core and a variety of functional groups on surfaces, which are often accompanied by the doping of heteroatoms including B, N, P and S[6–10]. Compared to traditional rare earth luminescent phosphor, CDs feature bright luminescence, ease of preparation and surface-functionalization, good biocompatibility as well as low cost and low toxicity. These characteristics make CDs significant in the application fields involving biology[11–15], chemical sensing[16–18] and optoelectronics[19–21]. Although CDs have been catching considerable attention as light sources for WLEDs and great progresses have been made so far, solid-state CDs still face enormous challenges as light-emitting materials due to the aggregation-caused quenching (ACQ) effect[22]. Therefore, currently numerous efforts have been devoted to inhibit the ACQ effect and enhance the optical properties of light-emitting devices, as may greatly promote the development and application of CDs in WLEDs.

Recently, although a few reviews have summarized the preparation, luminescence mechanism and application of CDs[23–25], the realization of solid-state fluorescence of CDs and their applications in WLEDs have seldom demonstrated in detail. Herein, we briefly overview the recent advances in the solid-state

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photoluminescence of CDs, and highlight their applications in WLEDs, particularly focusing on the current problems and future prospects. Typically, in the first section, a brief introduction to the synthesis and preparation of CDs is addressed, secondly followed by the demonstration of photoluminescence mechanism of CDs and the solid-state photoluminescence. In the third section, the applications in WLEDs were demonstrated in detail. Finally, the current challenges of solid-state CDs in WLED applications and the feasible perspectives were presented as well.

## 2　Synthesis of CDs

The morphology, size and degree of carbonization of CDs are intimately influenced by preparation methods. Accordingly, any difference in structure may significantly affects the performance of CDs in applications. Therefore, preparation techniques of CDs are critically important. Since CD was discovered, researchers have been pursuing simple, efficient and large-scale technique to prepare CDs of high-quality. Specifically, two types of strategies have been developed to obtain CDs, namely “Top-down” and “Bottom-up” approaches as shown in Fig. 1 (a-c refers to “Top-down” and d-f refers to “Bottom-up”).

"Top-down" means to cut and destroy large graphene structures of graphene oxide (GO), graphene nanosheets, carbon nanotubes (CNTs) etc. by either physical[26] or chemical techniques to obtain CDs. The methods involved, such as arc discharge[5,27], laser ablation[28–31], electrochemical synthesis[32–36] and so on, could efficiently transform large carbon structures into CDs. However, the commonly used advanced equipment or unique technologies result in higher costs of CDs. CDs synthesized by the “Top-down” strategy possess excellent graphene structure, but the surfaces are less functionalized with chemical groups. As a result, the π-π stacking interaction may occur between these CDs, which in turn leads to the decrease of fluorescence efficiency.

While the “Bottom-up” refers to the approach to obtain CDs by pyrolysis or carbonization of carbon-containing precursors upon chemical treatments. The carbon sources of the “Bottom-up” approach are extensive, ranging from small organic molecules or oli-

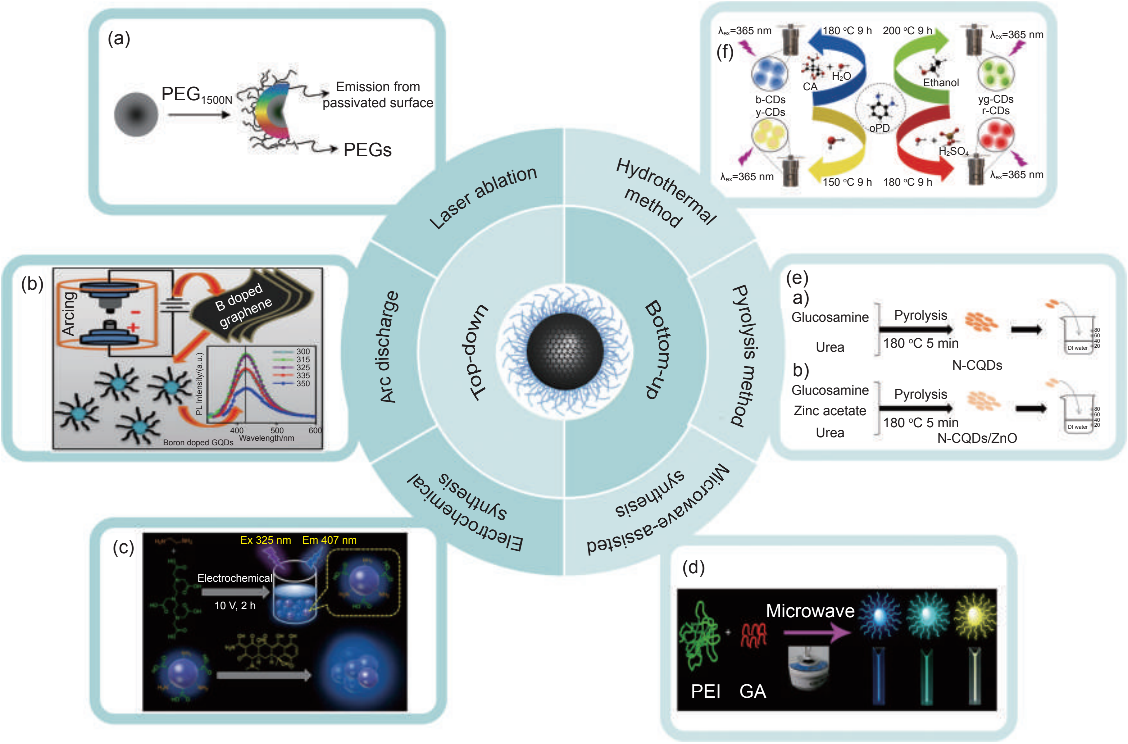

Fig. 1　Approaches to prepare CDs: “Top-down” and “Bottom-up”, (a) laser ablation[28], (b) arc discharge[27], (c) electrochemical synthesis[33], (d) microwave-assisted synthesis[47], (e) pyrolysis of precursors[54], (f) hydrothermal method[39]. (Reprinted with permission)

gomers of citric acid, urea, polyethylene glycol to carbon-enriched precursors of tea, orange peel and others biomass materials. The mostly used techniques include hydrothermal/solvothermal method[37–44], microwave-assisted synthesis[45–49] and pyrolysis method[50–55], etc. Compared to the “Top-down” strategy, CDs prepared by the “Bottom-up” feature a higher quantum yield (QY), a wider source of precursors, and lower costs. In particular, the hydrothermal/solvothermal method is regarded as one of the most simple and inexpensive technique. In these techniques, the precursor is dissolved in water or organic solvent and placed in a specially designed reaction vessel. The following carbonization of the precursor is accomplished under high temperature and pressure conditions. By optimizing precursor species, solvent, and the reaction parameters of temperature and time, the composition and PL color of CDs can be well regulated. The CDs synthesized by “Bottom-up” approach possess more surface functional groups, so a higher QY can be achieved. However, the as-received materials are often accompanied by many by-products in the synthesis process. The subsequent post-purification procedures become necessary to improve the fluorescence performance.

## 3 Photoluminescence mechanism of CDs

Excellent fluorescence emission is the most inherent and fascinating optical property of CDs. However, the PL mechanism of CDs is not completely clear so far. But with the advancement in CDs research, three PL mechanisms have been confirmed by researchers[56]: the size-dependent emission (determined by the carbon core)[57–58], the surface defect state emission (determined by the carbon backbone and the attached chemical groups)[59–62], and the molecular state emission (determined by fluorescent molecules linked on the surface or inside the CDs)[63]. These indicate that CDs exhibit a more complex system than they are expected.

### 3.1 Size-dependent emission

In general, the size of CDs (<10 nm) is comparable to that of traditional quantum dots (QDs). So some of researchers considered that the luminescence emission of CDs has the same size-dependent effect as that of traditional QDs. For CDs with perfect graphite structure, the size-dependent emission is mainly influenced by the carbon core. The energy band gap would decrease with the enlargement of carbon core, leading to a gradual red-shift of the emission peak.

The core of CDs has a polymer-like structure consisting of $sp^3$ hybridized amorphous carbon and small $sp^2$ hybridized atomic domains of various geometries. The $\pi$-$\pi^*$ transition on the aromatic ring accounts for the fundamental source of fluorescence emission. Density functional theory calculation studies show that the band gap width of the $\pi$-$\pi^*$ transition is related to the number of aromatic rings. When the number of benzene rings increases from 1 to 12, the band gap width of CDs decreases from 7 to 2 eV[57]. Therefore, there is a linear relationship between the emission wavelength of fluorescent CDs and their size. That is, the larger the size, the longer the emission wavelength. Yuan et al.[58] reported a size-dependent CDs of which the band gap width and emission wavelength are linearly related to the particle size, and the fluorescence lifetime of CDs decays single exponentially. These conclusions indicate that the fluorescence emission only comes from carbon core emission. Although the PL of CDs has experimentally proved to correlate with the size to some degree, this is not applicable to all studies of CDs. Therefore, PL mechanisms including surface-state emission and molecular-state emission are being explored.

### 3.2 Surface state emission

So far, the most common PL mechanism might be the surface-state emission for which a fluorescence emission center is formed *via* the synergistic hybridization of chemical groups with carbon core. In 2006, Sun et al. proposed a PL mechanism in CDs determined by the surface state[28]. They used $PEG_{1500N}$ to passivate the surface of CDs prepared by laser ablation approach, which demonstrated that the organic molecules could provide surface passivation and the

PL mechanism was controlled by surface defects. It is well known that the surface of CDs is rich of functional groups, and the so-called surface states are not composed of isolated chemical groups, but of the hybridization of carbon backbone and the attached chemical groups. The functional groups exhibit different energy levels, which may lead to a series of emission potential wells. The surface state emission potential wells would dominate the emission when light of a certain excitation wavelength irradiates to CDs. A higher degree of surface oxidation or other effective heteroatomic modifications would lead to more surface defects, which would result in an emission redshift. Oxygen-containing functional groups on carbon core are the main surface states of CDs, and the different surface oxidation accounts for different colors of fluorescence emission.

### 3.3　Molecular state emission

Another PL mechanism of CD could be attributed to the molecular state emission for which a fluorescence emission center is formed *via* organic fluorophores alone, where the fluorophores are attached to the surface or inside the carbon skeleton, leading directly to the emission of fluorescence. According to this theory, Song et al.[63] systematically studied the PL mechanism of CDs prepared from citric acid and ethylenediamine at different hydrothermal temperatures (Fig. 2). When the reaction temperature is low, citric acid and ethylenediamine are polymerized to form chromophores in the reaction system. As the system reaction temperature increasing, the molecular chromophore is further cross-linked to form a system in which cross-linked polymer dots and chromophore coexist. At this time, the luminescence of CDs is dominated by molecular states. As the temperature increases, the cross-linked polymer is further carbonized to form a carbon nucleus, and the chromophore in the reaction system gradually decreases. Accordingly, the proportion of molecular state luminescence gradually decreases. As a result, the carbon core state luminescence gradually dominates. CDs emitting through the molecular state exhibit strong fluorescence emission and high QY, while CDs emitting through the carbon core show better fluorescence stability.

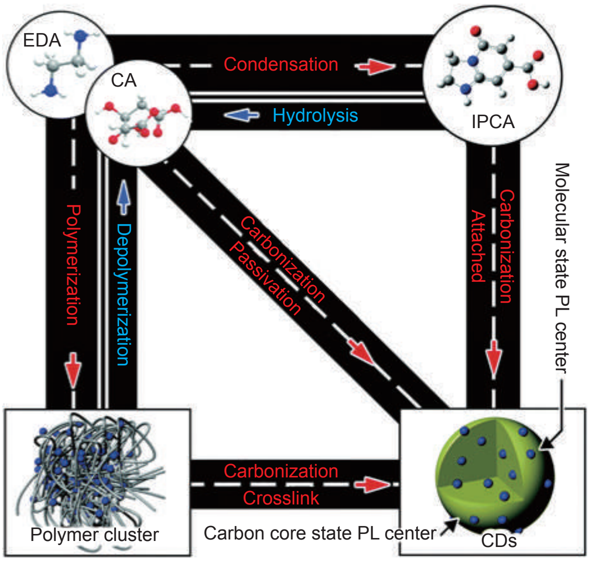

Fig. 2　A schematic of the relationship between different products in the one-pot hydrothermal system of CA and EDA[63].
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### 3.4　Strategies for fluorescence regulation

Heteroatomic doping is considered to be an effective method to improve the optical properties of CDs by adjusting the carbon skeleton matrix, chemical structure and energy gap[64]. Common elements such as nitrogen, sulfur, oxygen, boron, phosphorus and silicon are doped into CDs to adjust their optical properties and improve QY.

As one of the most commonly doped element, nitrogen (N) atom possesses the similar electron structure to that of carbon atom, and the unpaired electrons in nitrogen atom can be used as electron donors to improve the optical properties of CDs. For instance, Zhu et al.[65] demonstrated that if the precursor contains only —OH and —COOH groups, its PLQY is always less than 10%. However, if one or more amino groups are present, PLQY can exceed 10% or even reach a maximum of 80%. There are reports that proved the N contents and species of selected precursors are important in determining the optical properties of CDs[66]. Holá and co-workers[67] doped CDs with different types of N and got different colors of CDs, ranging from blue to red. In addition, the co-doping of two or more heteroatoms on CDs is becoming more and more popular in regulating fluorescence and improving QY. Using thiourea as both N and sulfur (S) sources, N and S co-doped CDs with red emission were synthesized, and the maximum emission wavelength reached 610 nm[68].

In summary, long wavelength emission CDs with large Stokes shift (especially red light) and improved QY could be achieved by heteroatomic doping which introduces more coordination sites or defects. This strategy is of great significance in fluorescence regulation of CDs.

## 4 Solid-state photoluminescence of CDs

Due to inter-particle interactions, mostly CDs in the aggregated state may produce non-radiative transition modes, such as energy transfer, surface electron transition and $\pi$-$\pi$ interactions between carbon cores, which in turn cause ACQ effects[69]. Therefore, maintaining the high fluorescence intensity and stability even in the solid state is a major challenge for CDs to be applied in WLEDs. In the past decade, the focus of CDs research has gradually shifted to the synthesis and application of solid fluorescent CDs. Typically, two strategies are employed to prepare solid-state fluorescent materials based on CDs. One strategy is to prepare CDs/matrix composites by mixing CDs with matrix, which is one of the most common routes to achieve solid-state fluorescence of CDs. Specifically, coating method and film-forming method account for preparing CDs/matrix based composites of phosphor and fluorescent films, respectively. Another strategy is to prepare the self-resistant quenching CDs to achieve solid-state fluorescence of CDs.

### 4.1 Coating method

Coatings have been widely studied as a major method for the preparation of solid-state photoluminescent CDs. The principle lies in that the precursor of CDs and the matrix of solid substrates are mixed and heated. As the precursor generates CDs, the matrix also starts to polymerize. During the polymerization process, CDs are wrapped to form a core-shell structure which enlarges the distance between CDs and avoids the mutual aggregation. Accordingly, the solid-state emitting of CDs is achieved. The common solid substrates reported in the literature include starch[70–72], polyvinyl pyrrolidone (PVP), polyvinyl alcohol (PVA)[73], silica gel[74], resin and some inorganic substances[75–76].

There are a large number of hydroxyl groups on their surface of starch granules, as could effectively absorb CDs. Moreover, the starch matrix neither competes for the absorption of excitation light nor affects the emission of CDs, so it was adopted previously for the preparation of solid-state CDs-based phosphors. Sun’s group successfully obtained starch/CDs phosphors with a QY of 50% by mixing starch with CDs in 2014. It showed great potential of CDs-based phosphors for LED, fluorescent signs and in other fields[70]. With the research progresses, Cao et al.[77] synthesized yellow-emitting CDs by a simple one-step hydrothermal method using o-phenylenediamine (o-PD) as raw material. Then, they developed a new environmentally friendly yellow CDs-based phosphorous with a QY of 66.9% by homogeneously mixing starch with CDs (Fig. 3(a)). Further, they used this phosphor as a single color converter to fabricate WLEDs with a color rendering index (CRI) of 83 and CIE coordinates of (0.342 9, 0.281 7), showing excellent performances (Fig. 3(b)).

In recent years, researchers have been devoting great efforts to CDs phosphors with better fluorescence performance and wider emission wavelength range. Aiming at this, Sun and his co-workers[78] achieved panchromatic emission of CDs by a solvothermal reaction of citric acid and urea at a constant mass ratio but by varying the concentration of the reactants in the solvent. Subsequently, they dispersed CDs chemically in $SiO_2$ network to produce effective full-color emitting $SiO_2$/CDs composites, of which the possible formation mechanism is shown in Fig. 3(c). When the $SiO_2$/CDs composites were applied to WLEDs, the desirable performance was achieved with a CRI up to 97.4. As experimentally confirmed, the choice of matrix materials is also important for the performance of CDs phosphors. In 2021, Cao et al.[79] reported the preparation of CDs phosphors using cal-

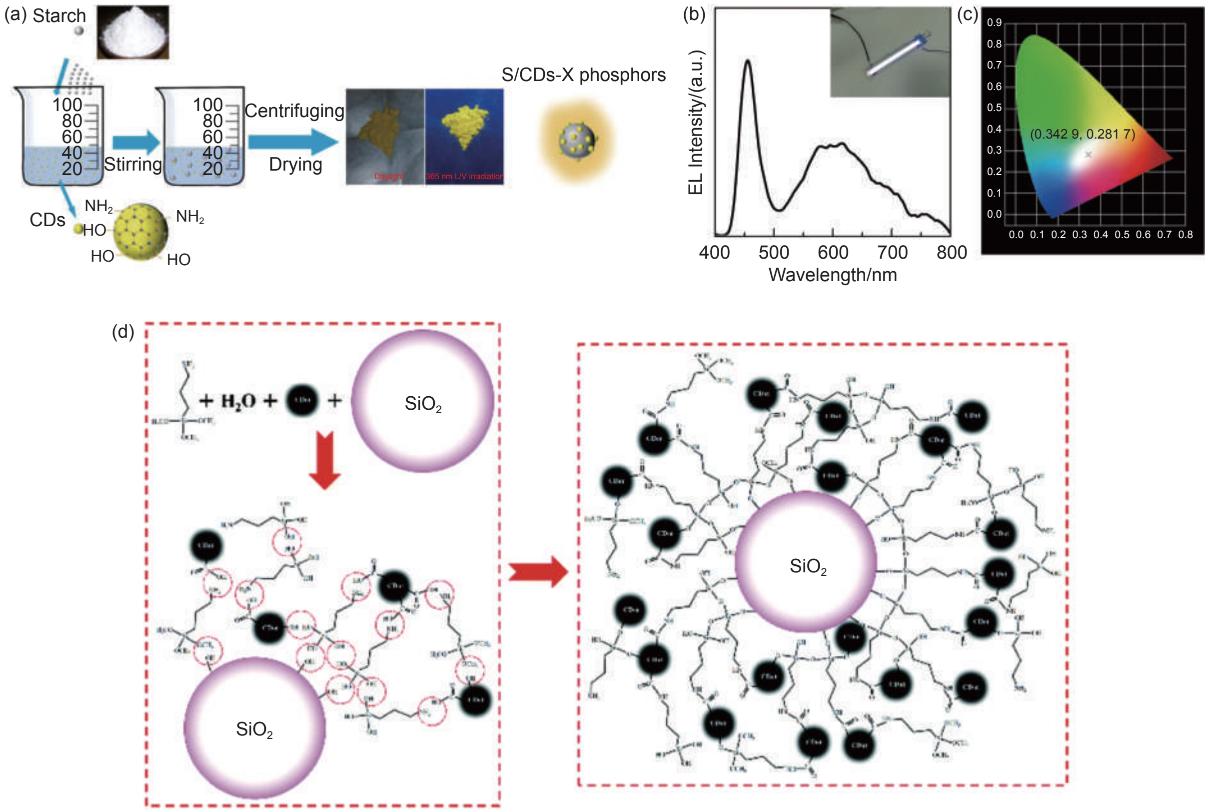

Fig. 3　(a) Schematic diagram of formation process for CDs phosphors based on starch, (b-c) the EL spectra, photograph and CIE coordinates of as-fabricated white LED operated at 30 mA current[77]; (d) possible formation mechanisms of the $SiO_2$/CDs composite phosphors by chemical dispersion[78]. (Reprinted with permission)

cium hydroxide ($Ca(OH_2)$) as a matrix material (Fig. 4(a)). The addition of $Ca(OH_2)$ could not only prevent the ACQ effect, but also further promotes the red emission of CDs, due to the abundant functional groups on the surface. Therefore, as shown in Fig. 4(b), the CDs phosphors prepared by mixing the green-emitting CDs with the $Ca(OH_2)$ exhibited green/red dual-emitting fluorescence (GRCDs) properties. It could be seen from Fig. 4(c, d) that GRCDs phosphors set for three months still maintain a stable morphology and structure. Thermal stability test proved that the phosphors synthesized using this method display excellent fluorescence and thermal stability (Fig. 4(e-g)). This discovery provides a new way for the development of novel CDs-based phosphors.

### 4.2　Film-forming method

Although coatings are effective to inhibit the ACQ effect to some extent and achieve solid-state photoluminescence of CD, low yields and complicated preparation procedures are unavoidable. Therefore, the direct film formation method has attracted the attention of researchers. This method introduces CDs into polymers (PMMA, PVA, PVP, or epoxy resin etc.) and obtains CDs-based luminescent films by the conventional method of preparing thin films.

In 2017, Miao et al.[37] reported a method to synthesize CDs with multiple color emissions by controlling graphitization and surface functionalization, and the emitted light covered the entire spectra (Fig. 5). In addition, they homogeneously dispersed CDs in epoxy resin to make CDs/epoxy resin composites. The as-prepared composite showed excellent performance in LEDs (Fig. 6). This research opens a new door for the development of low-cost CDs films to replace phosphors in light-emitting devices. In 2021, Lee and co-workers[80] prepared CDs capable of curing epoxy monomers, so that there was no need to add curing agents during the synthesis of CDs/epoxy composites, further simplifying the preparation scheme. The obtained CDs films retained more than 80% of the PL intensity after being stored at room temperat-

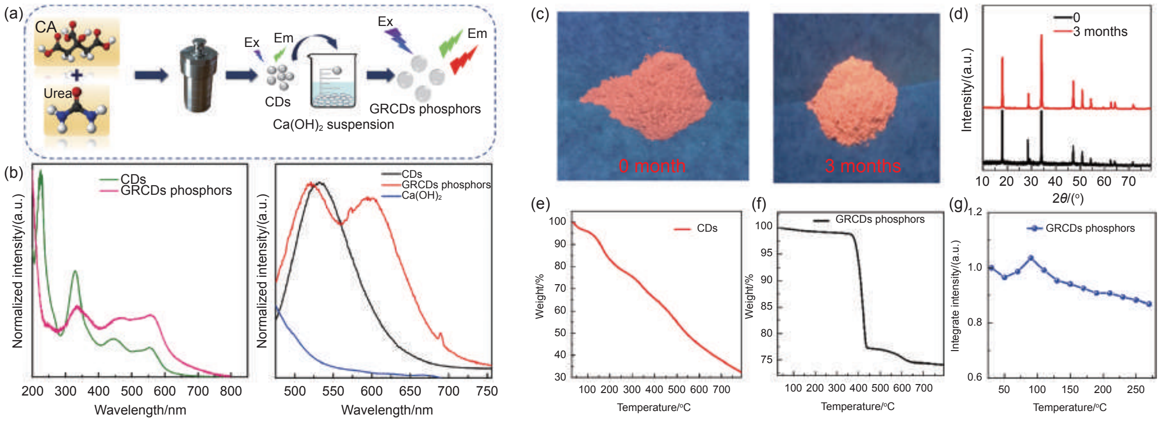

Fig. 4 (a) The synthesis process of GRCDs phosphors, (b) the absorption spectra of CDs and GRCDs phosphors and PL spectra of CDs, $Ca(OH)_2$ and GRCDs phosphors under excitation of 420 nm, (c-g) the fluorescence stability and thermal stability of GRCDs phosphors[79]. (Reprinted with permission)

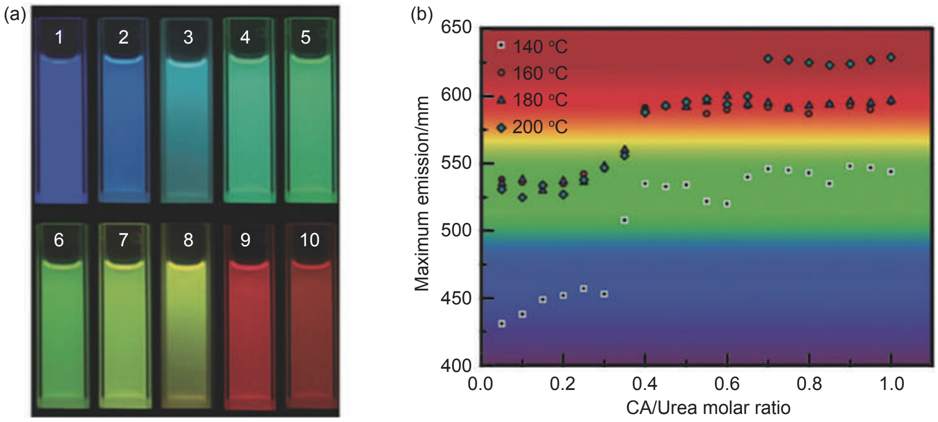

Fig. 5 (a) Optical images of luminescence CDs prepared from different reaction conditions under different excitation light, (b) the maximum emission peaks of CDs at different molar ratios of CA to urea and different reaction temperatures[37]. (Reprinted with permission)

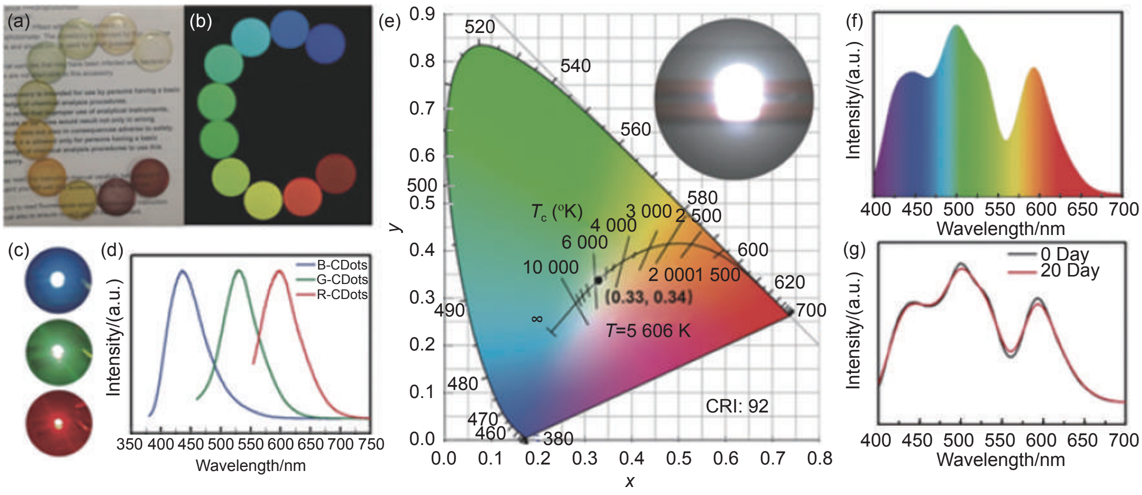

Fig. 6 (a-d) Photographs of CDs/epoxy composites and their application in LEDs, (e-g) performance and stability of WLEDs fabricated with CDs/epoxy composites[37]. (Reprinted with permission)

ure for 8 weeks. Recently, a transparent film of CDs/cellulose capable of efficient UV and high-energy blue light blocking was prepared by Barmon and colleagues[81]. Mou et al. developed a CDs/PVA composite film and used for antimicrobial and wound healing[82]. Kumari’s group prepared CDs fluorescent films with red, orange and yellow luminescence using PMMA and PVA as matrix materials, showing excellent performance in application of WLEDs[83]. These studies have demonstrated that CDs-based luminescent films exhibit excellent performance in photoelectronic devices. It is worth to believe that CDs could be utilized in more fields in future.

### 4.3　Self-resistant quenching CDs

Although it is possible to achieve solid-state luminescence by dispersing CDs in the matrix, this strategy suffers from a very low load rate of CDs in the solid matrix, and further increasing the load rate of CDs is always accompanied by a significant reduction in emission intensity. Therefore, the ACQ problem of CDs has not been completely solved yet, and the fundamental way to solve ACQ is to synthesize CDs capable of self-resisting fluorescence quenching. According to the luminescence mechanism of CDs, the luminescence is mainly regulated by carbon core and surface state. The optical properties of CDs are intimately dependent on the energy level structure caused by different graphitic structure of carbon core and surface groups[84]. Correspondingly, an effective way to inhibit the effect of ACQ lies in the regulation of the carbon core and surface structure of CDs.

#### 4.3.1　Regulation of carbon core

Generally, in CDs whose fluorescence emission is dominated by carbon core, the $sp^2$ hybridization domain directly determines the fluorescence characteristics of CDs. However, the existence of excessive $sp^2$ hybridization domain makes the π-π interaction of CDs in the aggregation state stronger, leading to non-radiative transition and fluorescence quenching. Therefore, reducing the $sp^2$ hybridization domain in carbon core could weaken the π-π interaction between particles, and then inhibit the ACQ effect. Li et al.[85] reported the preparation of high yield and anti-quenching solid CDs by microwave-assisted pyrolysis using potassium hydrogen phthalate (KHP), sodium azide ($NaN_3$) and boric acid (BA) as precursors. Compared with CDs solution, the excitation and emission spectra of solid CDs powder did not change much at all, maintaining relatively high QY (67.7% in solid and 73% in liquid). Notably, the structural characterization shows that there are a large number of $sp^3$ structures in the as-synthesized CDs, as greatly weakens the π-π interaction and inhibits the ACQ effect. This kind of diamond-like CDs with $sp^3$ structure provides a new solution to avoiding ACQ effect effectively (Fig. 7(a)).

In addition to reducing the proportion of $sp^2$ hybridization domain, the synthesis of CDs with uniform particle size also helps to suppress the ACQ effect. According to the size-dependent emission mech-

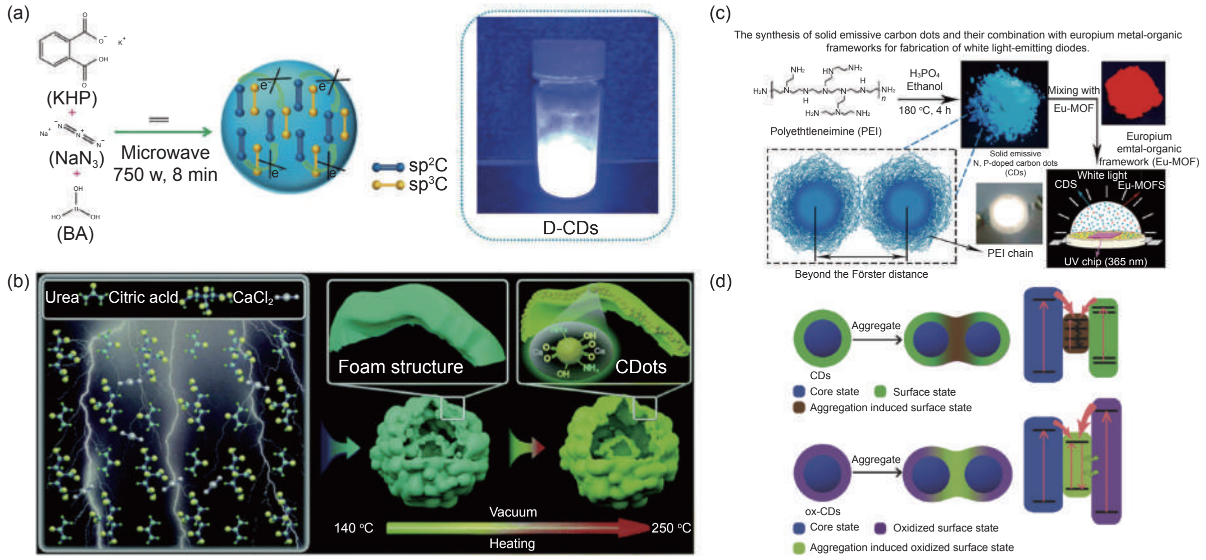

Fig. 7　(a) The structure of diamond-like CDs and a photograph under UV light[85], (b) schematics of the growth mechanism of CDs synthesized *via* the vacuum heating method[86], (c) the synthesis of solid emissive CDs and their application in WLEDs[87], (d) schematic of CDs and ox-CDs in dispersed and aggregated states, frames on the right show possible band-energy structures and quenching processes of CDs in the aggregated state and recombination processes of ox-CDs in the aggregated state[89]. (Reprinted with permission) anism, the conjugation size of carbon core determines the absorption/emission peaks. Therefore, in the CDs system with a wide distribution of sizes, the emission band gap differs in carbon core sizes. And energy transfers are more likely to occur during aggregation of CDs particles, leading to fluorescence quenching. Zhou and co-workers[86] developed a space-constrained vacuum heating method to obtain uniformly size-distributed CDs with high resistance to self-quenching. In a typical process, a mixture of citric acid, urea and $CaCl_2$ forms an inflated foam that was heated in a vacuum environment at 120 °C. The blue emissive molecular species were first formed when the temperature was gradually increased to 250 °C, and then transformed into uniformly distributed green luminescent CDs in the confined space of the foam through dehydration and carbonization process (Fig. 7(b)). The TEM and AFM images showed that the purified CDs had uniform particle size. These CDs exhibit bright green light emission in ethanol solution with QY as high as 72%, while maintaining QY as high as 65% in the solid state. The results show that only one luminescent center exists in these nanoparticles due to uniform particle size, which avoids the energy transfer between CDs in the aggregated state and effectively prevents the occurance of the ACQ.

### 4.3.2 Regulation of surface structure

Due to the existence of abundant surface functional groups, the surface interaction between nanoparticles in the aggregated state of CDs is an important factor affecting their solid-state luminescence. Accordingly, the inhibition of ACQ effect could be realized by regulating the properties of CDs surface functional groups.

One of the commonly used surface regulation strategies is to synthesize CDs with polymer chains structure on the surface using polymers as precursors. Polymer chains are able to cover around the carbon core, effectively extending the distance between neighboring particles and avoiding π-π interactions. Han et al.[87] designed a self-quenching-resistant CDs using polyethylenimide (PEI), phosphoric acid and ethanol as raw materials (Fig. 7(c)). The HRTEM images showed that CDs did not exhibit a significant lattice fringe, indicating that the precursors of PEI were not completely graphitized and a part of branched PEI chain covered the amorphous carbon core as a surface group. This structure increases the distance between adjacent particles, overcomes fluorescence quenching during aggregation, and endows CDs bright blue fluorescence in the solid state without the need for any other matrix. Chen et al.[88] firstly developed used hydrothermal method using PVA as carbon source and EDA as N source to synthesize green light emission CDs with emission wavelength of 525 nm in solid state, and the QY was up to 35%. Subsequently, a series of N-doped solid-state luminescent CDs were prepared by using different N sources. The emission wavelength varied to the different N content, and the long chain-like structure of PVA on the surface prevented the direct contact of luminescent centers, thus inhibiting the ACQ effect significantly.

Besides, surface functionalization could be employed to chemically modify the CDs' surface, so as to inhibit ACQ effect. Zhou et al.[89] prepared CDs with blue light emission in solution by microwave-assisted heating citric acid and ammonia water, then treated the CDs with hydrogen peroxide to obtain CDs (ox-CDs) capable of resisting fluorescence quenching in the aggregation state. In the excitation wavelength range of 270-500 nm, the ox-CDs powder displays an excitation independent emission peak of near 520 nm, and the maximum QY reaches to 25%. It was demonstrated that the hydrogen peroxide oxidizes the CDs surface without changing the structure of the carbon core. As shown in Fig. 7(d), the oxidized surface states possess higher energy levels, and they are coupled with each other to form a new absorption band gap in the aggregated state, which realizes the green light emission of ox-CDs powder. In 2021, Tao and his co-workers[90] synthesized CDs with blue emission in liquid state and cyan emission in solid state by a simple hydrothermal method using sodium citrate and dithiourea as raw materials. The existence of $Na^+$ permits CDs surface with a large number of sodium carboxylate functional groups that may inhibit π-π stacking interaction, resulting in the obtained CDs with excellent luminescence in the solid state. It is worth noting that the QY is even higher in the solid state (29.2%) than in the liquid state (7.6%), which may be attributed to the surface state change caused by aggregation.

## 5 Application of CDs in WLEDs

Due to the overwhelming superiority of energy saving, safety, high brightness and long life, WLEDs are becoming predominant in light-emitting devices research[91]. Table 1 summarizes the recent research progresses of CDs in applications for WLEDs. As shown, based on the excellent fluorescence stability and luminescence tunability of CDs, a large number of CDs derivatives have been developed and applied to WLEDs to replace conventional rare earth materials and semiconductor quantum dots. Depending on the luminescence principle, WLEDs are generally divided into photoluminescent (PL) WLEDs and electroluminescent (EL) WLEDs.

### 5.1 Photoluminescent WLEDs

Photoluminescent WLEDs were fabricated using CDs as a light conversion material to cover the chip that produces the excitation light. The white light emission with different color temperatures could be achieved by tuning the thickness and composition of CDs phosphor materials.

WLEDs could be prepared either by mixing CDs having different emission wavelengths or by directly using white light-emitting CDs. Fig. 8(a-e) shows the preparation of Blue fluorescent CDs (B-CDs) and their application toward WLEDs with high CRI[92]. By combining the as-prepared B-CDs with the green and red emitting CdTe-QDs working as color converters, white light emission was achieved, and WLEDs devices were thus assembled. The CIE coordinates (0.38, 0.36) were close to the standard value of white light under the excitation of UV chips. Moreover, Jiang and co-workers prepared o-CDs, m-CDs, p-CDs tricolor CDs using o-phenylenediamine(o-PDs), m-phenylenediamine(m-PDs) and p-phenylenediamine(p-PDs) as precursors, respectively[94]. Flexible

**Table 1 Summary of research progress of CDs in WLEDs**

| Emission color of CDs | Preparation method of CDs | Quantum yield (QY) of CDs | Methods | Correlated color temperature（CCT） | CIE coordinates | Color rendering index（CRI） | Ref. |
|---|---|---|---|---|---|---|---|
| Blue | Plasma-induced | 6% | PL WLEDs | / | (0.38, 0.36) | 87 | [92] |
| Blue | Hydrothermal | 45% (CDs phosphors) | PL WLEDs | 2805-7786 K | (0.29-0.41, 0.33-0.36) | 85-96 | [93] |
| Full-color | Solvothermal | / | PL WLEDs | / | (0.33, 0.34) | / | [94] |
| Red | Hydrothermal | 53% | PL WLEDs | 3875 K | (0.39, 0.39) | 97 | [95] |
| Red | Solvothermal | 23% | PL WLEDs | 5610 K | (0.33, 0.33) | 92 | [96] |
| Blue, Green | Hydrothermal | / | PL WLEDs | 6428 K | (0.31, 0.34) | / | [97] |
| Blue, Yellow, Red | Solvothermal | 64%, 57%, 51% | PL WLEDs | 5643 K | (0.31, 0.29) | 87 | [98] |
| White | Solvothermal | 5%-13% | PL WLEDs | / | (0.31, 0.33) | 96 | [99] |
| White | Solvothermal | 36% | PL WLEDs | / | (0.37, 0.39) | 87 | [100] |
| White | Microwave | 23% | PL WLEDs | 6987 K | (0.30, 0.35) | 83 | [101] |
| Blue | Solvothermal | 5% | PL WLEDs | 7093 K | (0.30, 0.34) | / | [102] |
| White | Hydrothermal | 9% | PL WLEDs | 3723 K | (0.39, 0.37) | 91 | [103] |
| White | Solvothermal | / | PL WLEDs | 6009 K | (0.32, 0.35) | 97 | [108] |
| White | Pyrolysis | 17% | EL WLEDs | / | (0.40, 0.43) | 82 | [104] |
| Blue | Microwave | 41% | EL WLEDs | 7694 K | (0.29, 0.33) | 83 | [105] |
| Yellow | Solvothermal | 44% | EL WLEDs | 2683-11240 K | / | 60-80 | [106] |
| White | Solvothermal | 19% | EL WLEDs | 4000-5000 K | (0.35, 0.36), (0.38, 0.41), (0.40, 0.43) | / | [107] |
| Red | Solvothermal | 86% | EL WLEDs | 3365 K | (0.38, 0.31) | / | [110] |

Note: "/" means that the item was not reported in the publication

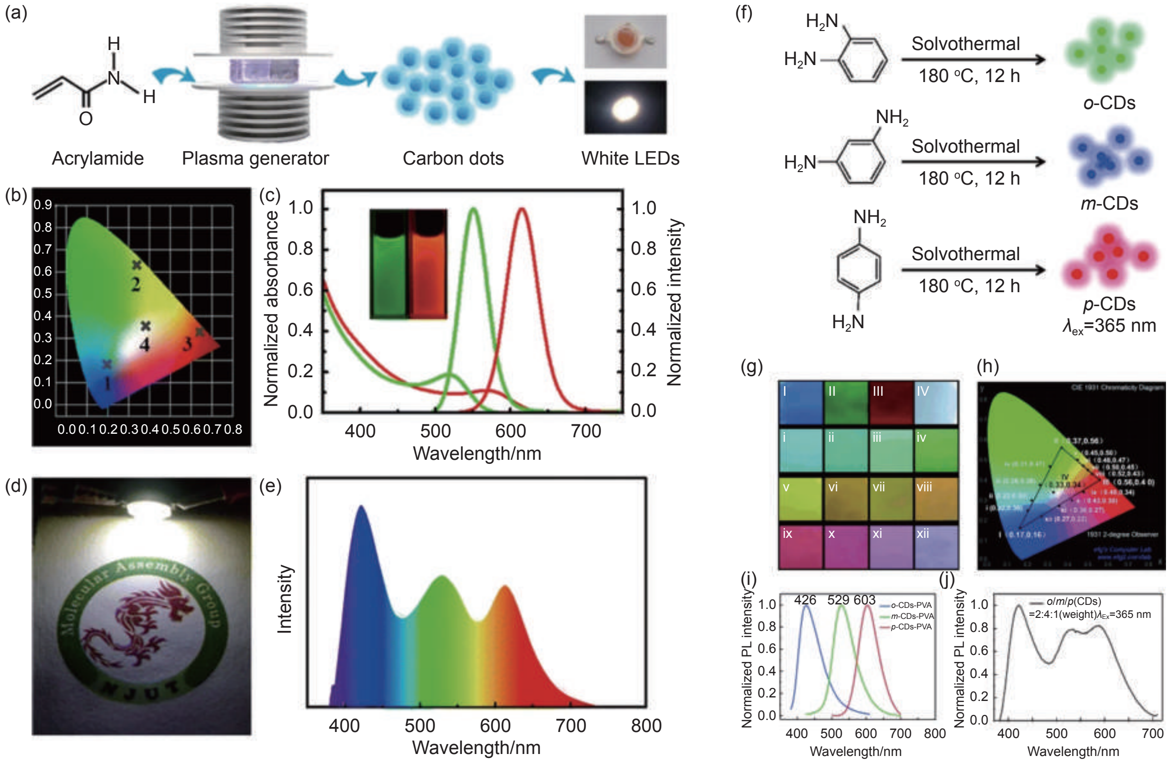

Fig. 8 (a-e) Synthesis of blue fluorescent CDs and their application in high color rendering index WLEDs[92], (f) schematic diagram of preparation of o-CDs, m-CDs and p-CDs, (g-j) Photographs of full-color CDs/PVA films under UV light and the performance of WLEDs fabricated with these films[94]. (Reprinted with permission)

full-color PVA films were prepared by mixing two or three of the aforementioned CDs in appropriate ratios. By further optimizing the mixing ratio of the CDs, white luminescent films with CIE coordinates of (0.33, 0.34) could be obtained (Fig. 8(f-j)).

In addition, with the synthesis of white light-emitting CDs being reported, researchers began to focus on the direct use of white CDs to prepare WLEDs. And it was shown that WLEDs prepared by using this strategy exhibited superior performance. Feng et al. prepared CDs with unique white emission in the solid state. By assembling them as phosphors with UV chips to form WLEDs, these devices exhibited satisfactory performance. Among them, the highest CRI reaches to 83, and the CIE coordinates (0.3, 0.35) are close to the standard white light coordinates[101] (Fig. 9(a)). Recently, Han and co-workers[103] reported the preparation of multicolor luminescent CDs and white luminescent CDs (W-CDs) using a single precursor (Fig. 9(b)). The CIE coordinates of W-CDs at the optimal concentration are almost same as the white light standard values (0.33, 0.33). Subsequently, as shown in Fig. 9(c), the WLEDs constructed from W-CDs/PMMA showed excellent performance, achieving a warm white light (CCT=3 723 K) with a CRI of 91.5 (>80 is excellent).

## 5.2 Electroluminescent WLEDs

Although encouraging progresses have been made in the study of photoluminescent WLEDs, safety problems caused by the leakage of ultraviolet light could never been completely solved. Alternatively, CDs with an electroluminescence (EL) property were developed to act as a light-emitting layer in the WLED structure. CDs-based electroluminescent WLEDs have a sandwich structure similar to that of quantum dot-based WLEDs, where CDs act as an intermediate active light-emitting layer surrounded by an interfacial transport layer and an electrode. In the typical structure of CDs-based electroluminescent WLEDs (Fig. 10), it could be divided into 5 parts, namely: anode, hole transport layer (HTL), active light-emitting layer (ALL), electron transport layer (ETL) and cathode. Among them, the ALL consists of CDs or CDs/polymer materials. When a voltage is ap-

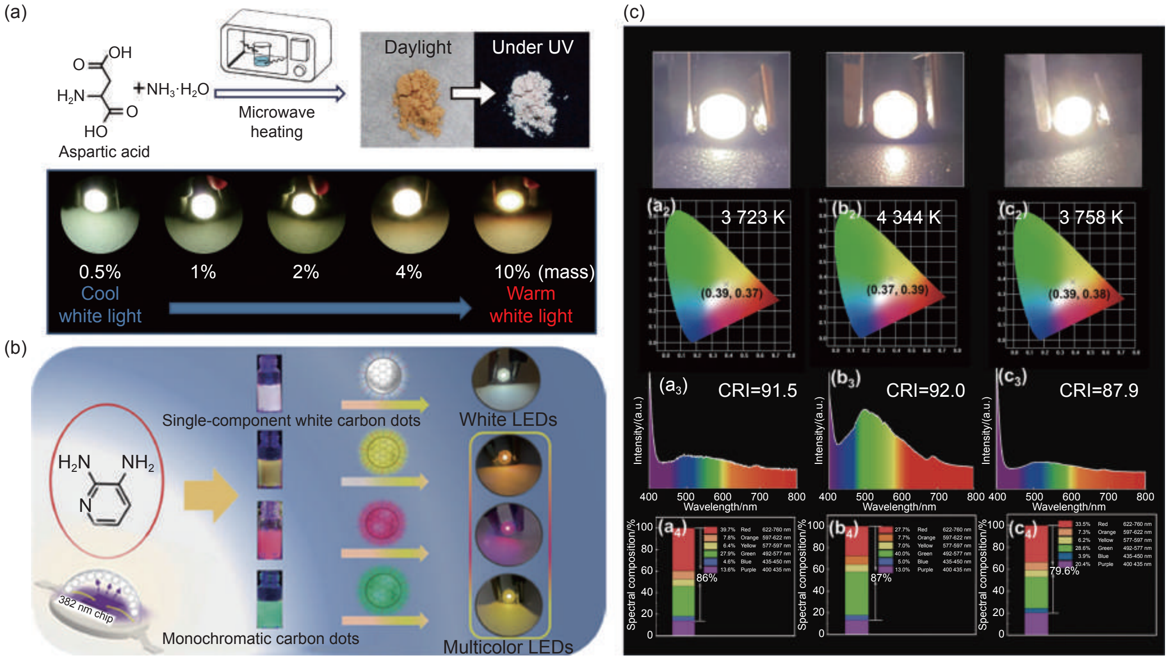

Fig. 9　(a) Synthesis route of W-CDs, photos of W-CDs under daylight and UV, and display diagram of WLEDs[101]; (b) Synthesis route of multicolor CDs[103]; (c) Construction of WLEDs with the W-CDs prepared in the cited article and performance of the devices fabricated[103]. (Reprinted with permission)

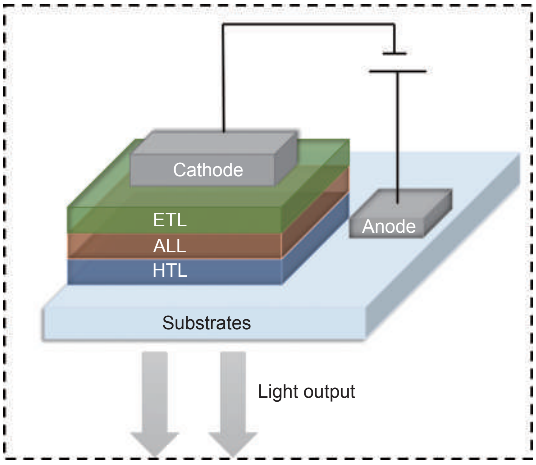

Fig. 10　Illustrations of the typical device structure of CD-based electroluminescent LEDs

plied to WLEDs, due to the applied electric field, holes and electrons are injected into the HTL and ETL, respectively. After migration, holes and electrons converge in the ALL and combine to produce excitons to trigger the light emission[109]. The luminescence of WLEDs could be modulated by changing CDs in the ALL.

In 2011, Wang and co-workers[104] reported WLEDs assembled from single-component carbon dots, which achieved a CRI of 82 at a current density of 5 mA/cm$^2$, comparable to that of some commercial WLEDs. The maximum external quantum efficiency reached to 0.083%, indicating the great potentials of CDs as white light electroluminescent devices (Fig. 11(a)). Later, Jia et al.[110] reported an electron-donating group passivation strategy to synthesize three types of red CDs with different emission wavelengths (Fig. 11(d)). The assembled WLEDs exhibited a maximum brightness of 5 248-5 909 cd/A and a current efficiency of 3.65-3.85 cd/A. The brightness still maintained over 80% of the initial value after 50 h of operation, showing excellent stability (Fig. 11(e-h)). Furthermore, a method capable of modulating WLEDs from cool to warm white light was developed by taking advantage of the luminescence redshifts and broadening caused by the aggregation of CDs, as accounts for the WLEDs with tunable CCTs from 2 863 to 11 240 K (Fig 11 (b-c))[106]. It is worth noting that these CDs-LEDs achieve a maximum luminance of 1 414-4 917 cd/m$^2$ and high external quantum efficiencies of 0.08%-0.87%. This work firstly demonstrated that CCT-tunable electroluminescent WLEDs could be obtained by controlling the aggregation of CDs. Recently, Zhou's group reported a route to synthesizing red CDs (R-CDs) and white CDs (W-CDs) by introducing free radicals. A record ex-

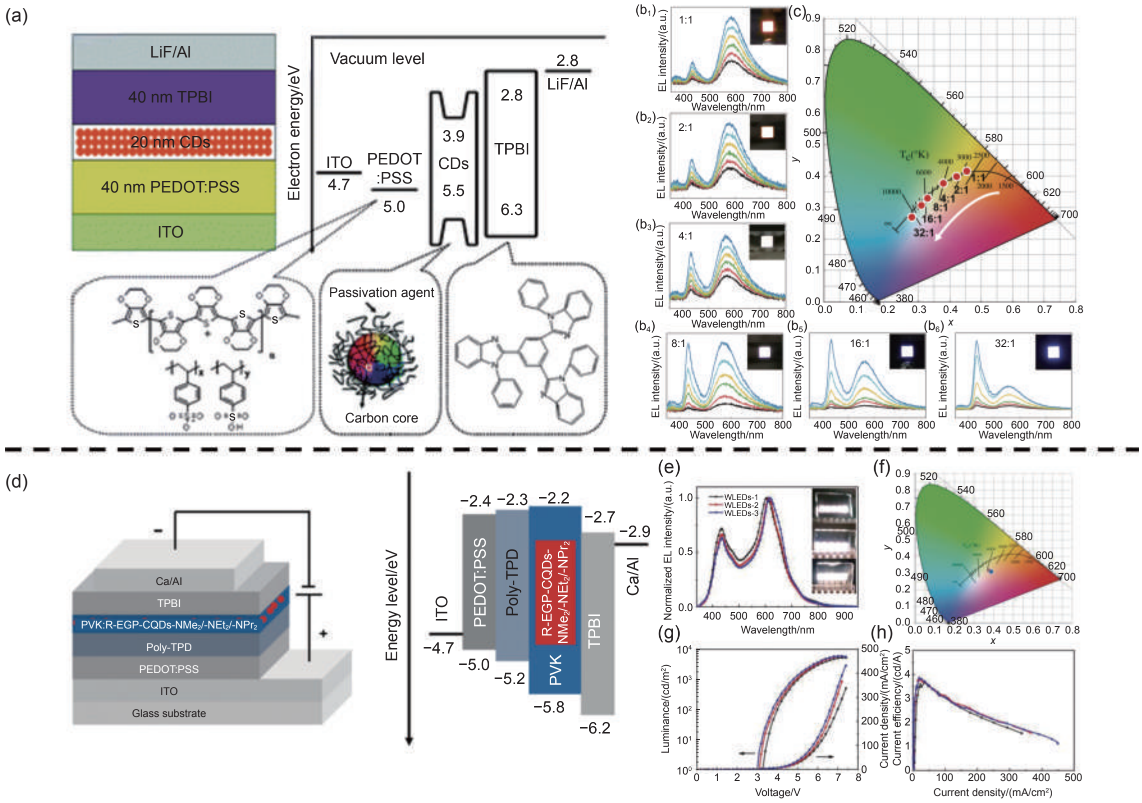

Fig. 11　(a) Schematic structure of electroluminescent WLEDs[104]; (b-c) Schematic of host/guest doping systems (PVK/Y-CDs and TFB/Y-CDs) together with the electroluminescence spectra and corresponding CIE coordinates of WLEDs prepared from films with different TFB: Y-CDs ratios[106]; (d) The device configuration and energy level diagram of WLEDs, (e-h) Performance test results of WLEDs[110]. (Reprinted with permission)

ternal quantum efficiency (0.95%) was achieved in the fabrication of electroluminescent WLEDs using W-CDs mentioned above[107].

Although fruitful achievements have been made with CDs in WLEDs, some of photophysical properties need to be deeply investigated to enhance the light stability, color stability, color rendering capability and luminous efficiency of CDs-WLEDs until they could be used on a large scale. In conclusion, CDs shows great potential in the fabrication of WLEDs, but more efforts should be devoted to achieve much lower-cost and higher-performance products.

## 6　Summary and outlook

Since the discovery of fluorescent CDs in 2004, the related research has been extensively conducted, due to their excellent emission fluorescence tunability, low toxicity, good biocompatibility and low cost. In this review, we summarized the main research progress of CDs in the aspects of preparation, photoluminescence mechanism, solid-state photoluminescence and applications in WLEDs in recent years. These progresses endow CDs with very broad application prospect and potentials.

Despite the great achievements on CDs, many challenges still remain. (1) The current existing mechanisms could only be effective for interpreting a given CD material, which makes it difficult to maximize the performance of CDs-based WLEDs. A universal and comprehensive explanation of the photoluminescence mechanism is strongly desired. (2) The research and development of solid-state luminescent CDs, especially the self-resistant quenching CDs, still face many challenges. The key point lies in the structural characteristics and quenching mechanism of CDs. More theoretical studies should be conducted to make the ACQ phenomenon determined by regulating carbon core and surface state explicit. In this regard, a universal method is desirable to be developed to design the structure of CDs and fundamentally solve the ACQ effect. (3) Although exciting achievements have been made in CDs-based WLEDs, these devices don't exhibit overwhelming performance as they are just expected over the commercial WLEDs. Particularly, the photo-physical properties of the CDs-based WLEDs need to be deeply investigated to enhance the light stability, color stability, color rendering capability and luminous efficiency, so as to be comparable in light-emission performances with rare earth materials based devices.

There is still a long way to commercialize the CDs-based WLEDs from laboratory to market. Fortunately, as CDs-based WLEDs are receiving more and more attention, we believe that these challenges will be overcome in the near future, and look forward to the explosive development of CDs in future lighting devices.

## Conflicts of interest

There are no conflicts of interest to declare.

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