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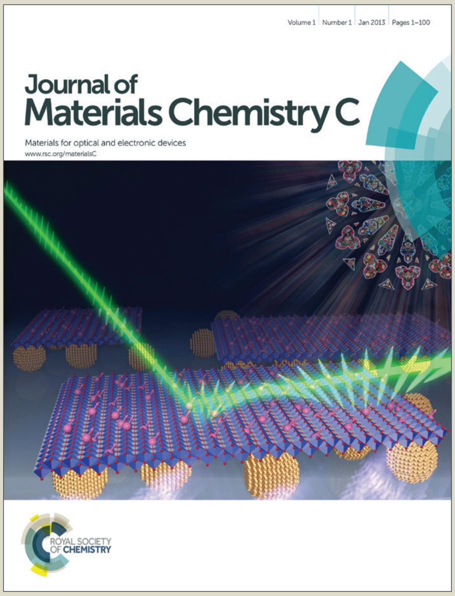

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TOC Summary and Graphic

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Fluorescent nanomaterial-derived white-light-emitting diodes: what's going on

In this review, we highlight recent progress of fluorescent nanomaterial-derived white LEDs, including semiconductor nanocrystals or colloidal QD-based LEDs, carbon-based LEDs, silicon QD-based LEDs, and organic-inorganic fluorescent nanocomposites derived white LEDs.

Journal of Materials Chemistry C

Cite this: DOI: 10.1039/c0xx00000x

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FEATURE ARTICLE

Fluorescent nanomaterial-derived white light-emitting diodes: what's going on

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Received (in XXX, XXX) Xth XXXXXXXXX 20XX, Accepted Xth XXXXXXXXX 20XX
DOI: 10.1039/b000000x

White light-emitting diodes (white LEDs) have recently attracted substantial interest owing to their remarkable energy conservation. The evolution of fluorescent nanomaterials with tunable optical properties has provided an opportunity for light source design of white LEDs. However, the stability and performance of fluorescent nanomaterial-derived white LEDs still fail to meet the requirements of practical applications. It is therefore imperative to boost their overall device performance, which depends on not only the exploitation of advanced fluorescent nanomaterials but also the design of superior light source. In this review, the achievements in fluorescent nanomaterials as color converters towards white LEDs are highlighted, including semiconductor nanocrystals or colloidal quantum dots (QDs), carbon-based nanoparticles, silicon QDs, and organic-inorganic fluorescent nanocomposites. The challenges and future perspectives in this research area are also discussed.

1 Introduction

Solid-state lighting in the form of white light-emitting diodes (white LEDs) has recently received worldwide attention, owing to their long lifetime and great economic significance.[1] The first commercially available white LED produced by Nichia Corporation was prepared by combining the blue InGaN diode chip with the yellow fluorescent inorganic $Y_3Al_5O_{12}:Ce^{3+}$ ($YAG:Ce^{3+}$) phosphor in 1996.[2] Since then, much effort has been devoted to improving the efficiency of white LEDs by developing novel color converters, which were used to convert the source light into the desired emission.[3] And the traditional rare-earth-based phosphors as color converters have played an important role in exploiting high-quality light source during the past few years. These phosphors, however, strongly depend on expensive rare earth resources with high energy consumption, and suffer from relatively low color quality.

Recently, white LEDs bearing internal active layers (inorganic and/or organic white emitting materials) have been investigated due to the advantages of the possibility to fabricate large-area, flexible and transparent devices *via* evaporation, co-evaporation and spin-casting techniques.[4-7] However, the purity of the color emission is strongly affected by the different ageing rate of the active layers, and the technological processes are complicated and expensive.[8] Comparatively, white LEDs based on fluorescent materials as color converters deposited on blue or UV diode chips can easily acquire high color quality.

To evaluate the quality of the white light emitted from white LEDs, several important parameters have been introduced: (i) color rendering index (CRI), (ii) correlated color temperature (CCT) and (iii) Commission Internationale de l'Eclairage (CIE) chromaticity coordinates. The CRI, ranged from 0 to 100, is a quantitative measure of the ability of a light source to reproduce the colors of a variety of objects in contrast to an ideal or natural light source.[9] A high CRI (>80) is required in color-critical applications especially indoor lighting.[5] The CCT is the temperature of a blackbody radiator emitting the same colour of the light source. The temperature in Kelvin (K) at which the heated black-body radiator matches the color of a tested light source is the color temperature of that source. In general CCT values for white LEDs have a range from warm white (2500 to 4500 K) to cold white (4500 to 6500 K).[10] The CIE chromaticity coordinates (x, y) maps colours visible to the human eye in terms of hue and saturation. For example, (0.33, 0.33) is the desired CIE coordinate value for white light.[4] Traditionally, the luminous efficiency (LE) of white light, reported as in lumen per watt (lm W^{-1}), represents the output light power from a device. The brightness is reported as either their external quantum efficiency (EQE) or in candelas per amp (cd A^{-1}).[11]

Ideal white LEDs should be highly efficient, cost competitive, and have a long lifetime, and hence high-efficiency light conversion materials are intensively required. Fluorescent materials, especially in the form of nanomaterials, have long been

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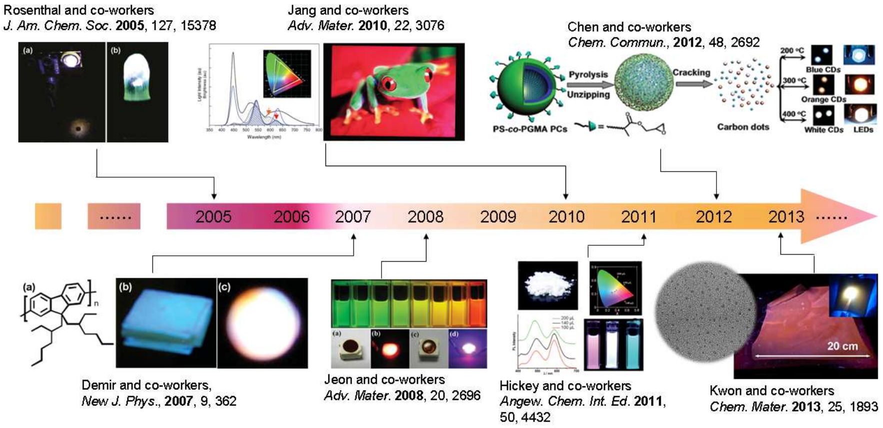

Fig. 1 Timeline showing recent progress in fluorescent nanomaterial-derived white LEDs in the literature. Reproduced from refs. [1, 12, 13, 14, 15, 16, 17].

researched as potential color converters for LEDs because of their ease of large-scale fabrication and abundance in nature. The size, morphology and orientation of fluorescent nanomaterials can also be easily modified, which enables us to systematically investigate the relationships between structure and optical properties. To date, although some fluorescent nanomaterials have been proposed to fabricate white LEDs, it still doesn't satisfy (usually photooxidatively and thermally unstability) the commercial standard (brightness of 3300-5500 cd m^{-2}, LE of 70 lm W^{-1} and 5,0000 hours of lifetime simultaneously). In order to address these issues and exploit high-efficiency white LEDs, more efforts are urgently needed on the development of novel fluorescent nanomaterials together with advanced devices architecture.

In this feature article, we would like to overview the recent progress in the development of fluorescent nanomaterials as conversion phosphors towards white LEDs. Several of the outstanding advances made to date are illustrated in Fig. 1. In the following sections, the paper is divided into three parts: (a) semiconductor quantum dots (QDs) based white LEDs, (b) group IV fluorescent nanoparticles based white LEDs (e.g., carbon-based nanoparticles based white LEDs and silicon QDs based white LEDs) and (c) organic-inorganic fluorescent nanocomposites based white LEDs.

2 Semiconductor QD-based white LEDs

Colloidal semiconductor nanocrystals or quantum dots (QDs) have unique and attractive characteristics such as narrow emission spectral bandwidth, high photoluminescence quantum yields (PL QYs), as well as size-tunable optical properties.[1, 3, 14, 18-27] In addition, excellent photochemical stability can be attained by intensive modification of the QD surface, favouring an improvement in stability and efficiency of the devices. These unique properties make QD a promising candidate as one of the most practical light converters for QD-based light-emitting diodes (QD-LEDs).[28] Ever since the first QD-LED was demonstrated in 1994,[29] the research on white QD-LEDs has rapidly increased. To date, much effort has been focused on optimizing the optical properties, improving the stability, enhancing the compatibility of QDs with the silicone, and reducing the toxicity of QDs. The following sections will show recent developments in the general method for the fabrication of stable white LEDs comprising QDs.

2.1 White LEDs based on blue InGaN chips and colloidal semiconductor QDs

There are three types of white LEDs: multichips white LEDs, ZnSe-based white LEDs, and single-chip InGaN white LEDs. Multichips white LEDs exhibit three emission bands and possess a good CRI by constructing a red-, a green-, and a blue-emitting chip. However, they suffer from high cost and need a relatively complicated external detector and feedback system because each chip degrades at a different rate.[30] ZnSe-based white LEDs can emit white light through blending blue light from a ZnSe active layer with yellow light from a ZnSe substrate,[31] but they have a lower emission efficiency and a shorter lifetime than InGaN white LEDs, therefore they are less studied in past decades. Single-chip white LEDs, which comprise only a blue InGaN chip and a yellow-emitting yttrium aluminum garnet (YAG) phosphor, will be used as general lighting in the future.[32] Although YAG doped with cerium (YAG:Ce) phosphor is most widely used in single-chip white LEDs, they have a low CRI because of their lack of emission in the red region. To address the issuse, the application of QDs as the downconverter in the white LEDs can serve as a promising alternative and play an important role in improving the CRI.

Demir and co-workers demonstrated tunable white light generation utilizing combinations of CdSe/ZnS (core/shell) QDs with the blue-emitting InGaN chips.[33-35] The size of the QDs determined the colors which contributed to white light generation. The PL intensity could be adjusted by the QD concentration and the QD film thickness of the color conversion layer and the transmitted electroluminescence (EL). As a result, the CRI of this device could be adjusted from 14.7 to 82.4, which displayed their widely tunable colour properties. Recently, Kim *et al.* reported a white LED with an improved CRI value of 85.0, which combines a blue LED with the blend of YAG:Ce phosphor and CdS:Mn/ZnS core/shell QDs in a weight ratio of 1:1.[36] Yellow emitting YAG:Ce phosphors and 4.5 nm-sized orange-emitting CdS:Mn/ZnS core/shell structured QDs were prepared by a

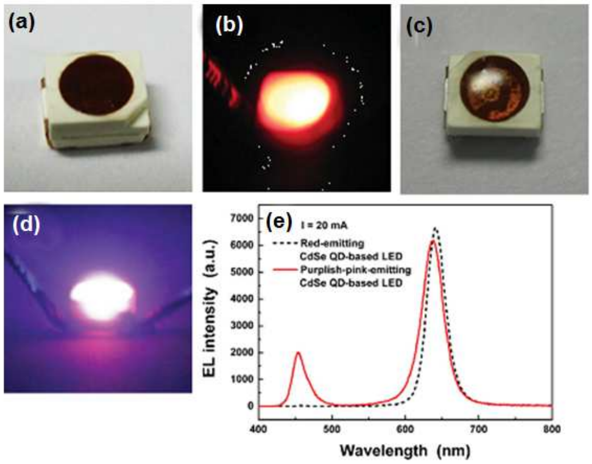

Fig. 2 Photographs of: (a) an as-prepared CdSe-based red LED, (b) a CdSe-based red LED in operation, (c) an as-prepared CdSe-based purplish-pink LED, (d) a CdSe-based purplish-pink LED in operation, respectively. (e) EL spectra of both CdSe-based LEDs when operated at 20 mA. Reprinted with permission from ref. 14. Copyright 2008 Wiley InterScience.

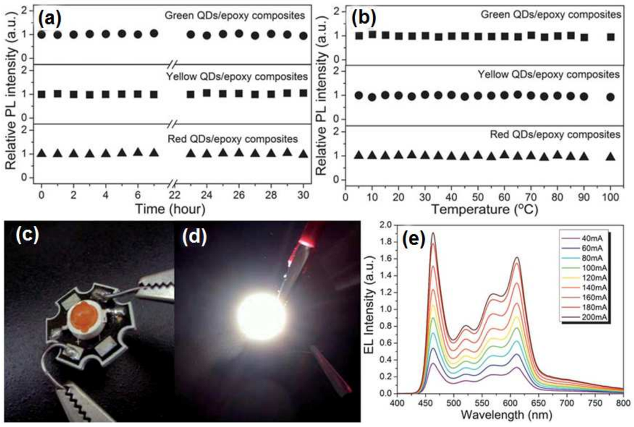

Fig. 3 Time-dependent evolution of the relative PL intensity of the CdSe/CdS/ZnS QDs/epoxy composites: (a) with three emitting colors under 365 nm UV lamp radiation and (b) at temperature ranging from 5 ºC to 100 ºC. (c) Digital pictures of the as-prepared white LED and (d) operated at 100 mA. (e) EL spectra of the as-prepared LED under different forward bias currents. Reprinted with permission from ref. 3. Copyright 2011 Royal Society of Chemistry.

modified polyol and a reverse micelle chemistry method, respectively. To compensate the poor CRI of YAG:Ce-based white LEDs due to the lack of red spectral component, CdS:Mn/ZnS QDs were blended into YAG:Ce nanophosphors. Blends of nanophosphors and QDs exhibited the prominent spectral evolution with an increasing content of QDs. In addition, various novel QDs such as CdSeS, $CdSe_xS_{1-x}$/ZnS and CdTeSe/ZnS have been loaded on blue LED chips to give excellent optical performance.[37-38] A recent development comes from Xu and co-workers, who fabricated novel water-based core/shell CdTeSe/ZnS QDs (QY = 16%) by aqueous method as the color convertor.[38] In this case, the CRI of the as-fabricated white LEDs can reach as high as 83.3. Following this similar concept, another white LED based on YAG: Ce,Gd phosphor and CdSe/ZnS QDs has also been fabricated.[39] In this study, white LED was achived by combining a blue LED with the blends of YAG: Ce,Gd phosphor and CdSe/ZnS QDs in a weight ratio of 1:1. As a result, the as-obtained white LED showed excellent white light with CIE coordinates of (0.33, 0.32), CRI of 90.0, and LE of 80.0 lm/W^{-1}, respectively.

To obtain high-quality white light, Jang and co-workers have reported a white LED with CRI of 90.1 and CCT of 8864 K by hybridizing CdSe QDs and Sr_3SiO_5:Ce^{3+},Li^+ phosphors on blue InGaN chips.[14] In this work, various emissive lights such as pink, purple, and yellowish-pink could be generated from CdSe QD-based LEDs *via* varying the concentration of CdSe QDs (Fig. 2a-d). Compared to the red-emitting CdSe QD-LED, the red emission of the purplish-pink LED shifted from 638 nm to 641 nm), and the bandwidth decreased by 4.2 nm (Fig. 2e). By replacing CdSe QDs with CdSe/ZnSe core/shell QDs, Jang and co-workers have also fabricated white LEDs with an CRI of 85.0 and a CCT of 6140 K.[40] The incorporation of red-emitting CdSe/ZnSe QDs into a greenish-yellow-emitting Sr_3SiO_5:Ce^{3+},Li^+ phosphor is advantageous for coverting a wide visible spectral range and obtaining higher CRI. The results indicate that the combination of phosphor and QDs in LEDs can be a good solution to obtain white light sources with high color rendering properties. Moreover, another novel nanocomposite blend of CdSe/CdS/ZnS red QD, Sr_2SiO_4:Eu green phosphor and silicone resin was demonstrated as a color converting material.[41] The white LED converted by this newly developed QD-phosphor nanocomposite produced light with CIE coordinates of (0.30, 0.33), device temperature of 94 ºC, CCT of 6672 K, LE of 65.9 lm W^{-1} and CRI of 83.2, which revealed that it can be used for high-power applications and is potentially suitable for use as a general illumination source.

To acquire the desired white LEDs, the thermostability and photostability of devices are also important. Under ordinary circumstances, the PL intensity of QD ensembles is a reversible function of the temperature.[42-43] With an increase in temperature, the PL intensity decreases and the spectral wavelength shifts towards red. Accordingly, amount of heat carried by the operating LED chips would affect the color quality and the liftime of white LEDs. Much effort has been devoted to tackling this issue in the past few years. The current approaches to enhance the stability mainly involve developing core/multishell QDs, process intensification of device, and designing new device structures.

The core/multi-shell system offers higher stability against photo-oxidation and higher QYs than the core/shell system. Up to now, various core/multi-shell QDs such as CdSe/ZnSe/ZnS, CdSe/ZnTe/ZnS, CdSe/CdS/ZnS, and CdS/$Zn_{0.5}Cd_{0.5}$S/ZnS have also been synthesized.[44-47] For instance, Wang *et al.* recently successfully synthesized CdSe/CdS/ZnS core/multi-shell QDs with high QY (~70%) and excellent optical stability *via* a phosphine-free and continual precursor injection method in paraffin liquid, which was used for white LEDs.[3] In this case, QDs/epoxy composites were then fabricated by directly dispersing the core/multi-shell QDs into epoxy resin as the encapsulating and light conversion material, and the as-prepared composites possessed even better stability against UV radiation and heating, as shown in Fig. 3a and b. Moreover, a white LED was then fabricated by combining blue InGaN chip and QDs/epoxy composites incorporated in green, yellow and red emission QDs, and the resulting fourband RYGB white LED showed good performance with a CIE coordinates of (0.35, 0.37),

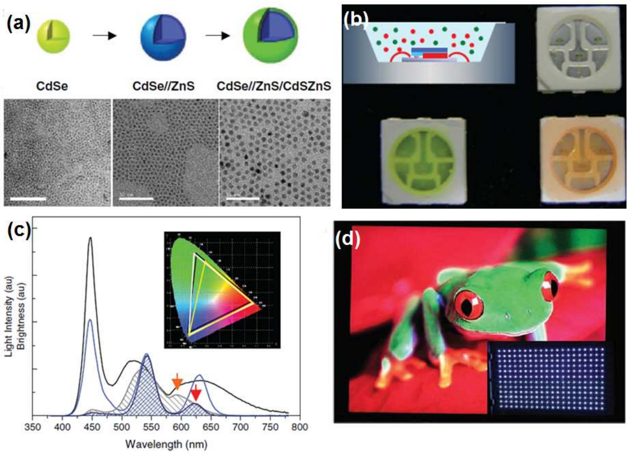

Fig. 4 (a) Schematic structures and TEM images of core/multishell QDs. (b) Schematic diagram of QD LED, blue LED package (*upper right*), green QDs (*lower left*) and red QDs with silicone resin dispersed and cured (*lower right*). (c) Light intensity spectra (*solid line*) and brightness (*hatched area*) of the QD-LED (*blue*) and the phosphor-LED (*grey*). Inset: Color triangles of the QD-LED (*white*) and the phosphor-LED (*yellow*) compared to NTSC1931 (*black*). (d) Display image of LCD TV with white QD-LED backlights. Reproduced with permission from ref. 1, Copyright 2010 Wiley InterScience.

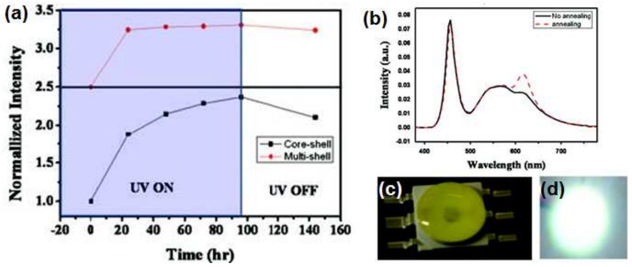

Fig. 5 (a) Time response of PL intensity of core/shell QD nanocomposites. (b) Luminance characteristics of LED before and after UV illumination. Digital pictures of the as-prepared white LED: (c) without operating and (d) and operating at s 350 mA. Reprinted with permission from ref. 24. Copyright 2011 Wiley InterScience.

CRI of 88.0, CCT of 3865 K and a LE of 32.0 lm W^{-1} at 100 mA (Fig. 3c-e). The results suggest that the incorporation of highly fluorescent CdSe/CdS/ZnS core/multishell QDs into LEDs can be a promising solution for white light sources with high color rendering and stability properties. In a recent work,[1] Jang and co-workers developed the highly luminescent, multishell-structured green CdSe//ZnS/CdSZnS QDs (Fig. 4a) and red CdSe/CdS/ZnS/CdSZnS QDs showing up to 100% PL QY. Subsequently, the as-prepared QDs were wrapped as green and red color converters in InGaN blue LEDs (Fig. 4b) to fabricate white LEDs for display backlights, as shown in Fig. 4d. And this as-prepared cool-white QD-LED (at the color coordinate of (0.24, 0.21)) showed 41.0 lm W^{-1} at a CCT of 100,000 K and more than 100% color reproducibility compared to the National Television Systems Committee (NTSC) standard in the CIE 1931 color space. Additionally, the EQEs reached 72% for green-light emission and 34% for red-light emission respectively, and the lifetime maintained above 2200 h under ambient conditions. More interestingly, the white QD-LEDs were successfully used as backlight for a 46 inch LCD panel for the first time.

It should be noted that thermal annealing and photoactivation techniques can also be used to boost the PL intensity of QD nanocomposites at specific conditions.[48-49] Although the QD nanocomposites often show PL diminishing or quenching at elevated temperature under normal circumstance.[42] Niu *et al.* developed a thermal annealing approach to improve the performance of QD LEDs.[48] After an *in-situ* annealing at 180 °C for the CdSe/CdZnS/ZnS QDs, the EQEs were increased by $\sim$3 times compared to identically prepared devices without thermal annealing. In addition, Kim *et al.* recently firstly demonstrated the irreversible PL enhancement *via* photoactivation.[24] In this study, a high-power UV lamp was utilized to improve the stability of device. As shown in Fig 5a, the PL intensity of two samples (CdSe/ZnS core/shell and CdSe/CdS/CdZnS/ZnS core/multishell nanocomposites) increased significantly and gradually reached a saturated value after 96 hours of UV irradiation. Exhilaratingly, after the UV lamp was moved, the PL intensity of core/multishell nanocomposites retained, revealing high photostability of these nanomaterials. The UV annealed white LEDs fabricated with core/multishell QD nanocomposite have improved both in terms of brightness and CRI (from 87.2 to 91.0) compared with no UV treated ones (Fig. 5b-d). The results showed the UV effect depends on two critical factors: UV intensity and exposure time. Particularly, another effort focuses on the development of heavy metal-free QD-based white LEDs, given the global trend to regulate use of environmentally friendly materials. Indium phosphide (InP, band gap: 1.35 eV) is regarded as the desired alternative fluorescent material, which provides a similar emission wavelength range and without intrinsic toxicity.[50-54] Recently, high PL QY and stable emission of InP-based QDs have been realized.[55-58] Kim *et al.* developed successive synthesis of InP/ZnS QDs employing an autonomous hybrid flow reactor *via* the combination of a batch-type mixer and a flow-type furnace.[59] In this way, different-sized InP/ZnS QDs could be synthesized in continuance and large scale

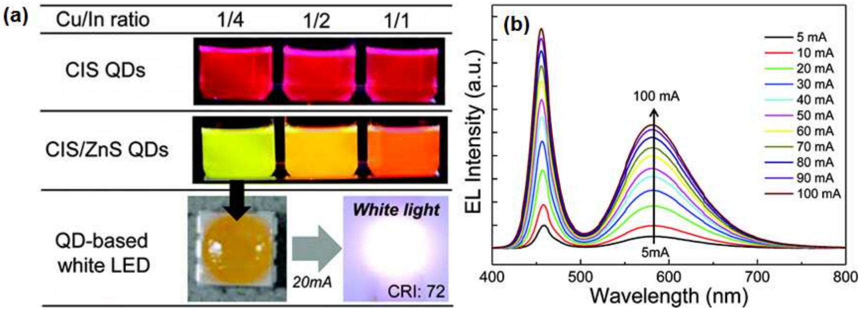

Fig. 6 (a) Schematic diagrams for synthesis of CIS and CIS/ZnS QDs and fabrication of CIS/ZnS QD-based white LED. (b) EL spectra of white LED operated at 5 to 100 mA. Reprinted with permission from ref. 62. Copyright 2012 American Chemical Society.

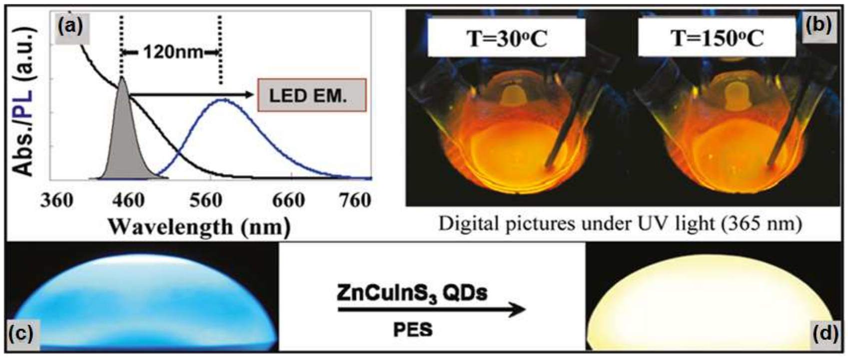

Fig. 7 (a) Spectra of CZIS QDs sample and LED emission at ~460 nm. (b) Digital pictures of CZIS QDs heated to the different temperatures under UV light (365 nm). Photographs of LEDs before (c) and after (d) being coated by the CZIS QDs. Reproduced with permission from ref. 63. Copyright 2011 American Chemical Society.

by controlling the flow rate and temperature of the reactor. Subsequently, a white LED was manufactured with a CRI value up to 89 by adding InP/ZnS QDs together with yellow phosphors, which was desirable for solid-state illumination applications. Otherwise, Ziegler *et al.* demonstrated an available method to achieve more stable InP/ZnS QDs *via* the passivation of surface defects with inorganic SiO_2 rather than with organic ligands.[60] The QDs' performance was increased to 50% QY after the surface modification through the microemulsion synthesis. Thus, the modified QDs were more beneficial to be used as colour converting materials for white LEDs applications.

Ternary-type QD white LEDs have been also successfully developed through various approaches in recent years. Among those reports, Cu-In-S (CIS) QDs are of particular interest owing to their stable absorption and emission features. Zhong and co-workers recently developed a high-quality CIS QDs (PL QYs of 60% for green emissive samples and 75% for yellow and red emissive samples) *via* a facile non-injection method.[61] By adding green and red emissive CIS QDs on InGaN-based chip, they explored two-typed white LEDs. The as-fabricated LEDs with surface mounted device types revealed excellent properties (CRI of 95.0, LE of 70.0 lm W^{-1}, and CCT of 4600-5600 K).

Impressively, another type of white LED (high-power type) also showed a high CRI of 90.0. It turned out that the nontoxic Cu-In-S QDs could be one of the candidates for commercial applications. Similarly, one white LED was built with Zn doped-CIS QDs but showed lower CRI.[64] Very recently, Yang's group successfully utilized a hot colloidal route to prepare the CIS/ZnS core/shell QDs with quite widely tunable emission colors (623 nm, 598 nm and 564 nm), as shown in Fig. 6a.[62] In this context, the as-obtained CIS/ZnS QDs were combined as color converters with a blue LED chip to fabricate white LED. As shown in Fig. 6b, with increasing forward bias, both blue and QD emissions increased. Most recently, a novel InP/GaP/ZnS core/shell/shell QD with QY (~85%) was first fabricated by *in situ* method towards white LEDs.[66] InP/GaP/ZnS QDs showed much superior thermal stability over InP/ZnS QDs reported in previous work,[67] ascribed to the efficient shell passivation. Herein, the GaP inner shell played an important role in mitigating the effect of lattice mismatch between the InP core and the ZnS outer shell.[66]

Quaternary QDs have also been exploited to fabricate white LEDs. Zhang *et al.* successfully developed quaternary Cu-Zn-In-S (CZIS) QDs, and the QDs' optical performance and digital pictures of LEDs are shown in Fig. 7.[63] A new synthetic approach was exploited to fabricate various homogeneous CZIS QDs by varying reaction temperature and the ratio of the precursors, respectively. This as-prepared QDs exhibited not only broad PL spectra with emission color tuned from visible to NIR region (Fig. 7a), but also high QY (over 70%) without coating the wide band gap shell materials. Further studies revealed good thermal stability, where the CZIS QDs did not show any obvious variation in emission intensity upon heating to 150 ºC by visual observation, as shown in Fig. 7b. Song *et al.* recently developed color-tunable Cu-In-Ga-S (CIGS) QDs and CIGS/ZnS core/shell QDs by adjusting the ratios of In: Ga to apply in white LEDs.[65] The EL spectra and digital pictures (Fig. 8b) of CIGS/ZnS QD-based LEDs with different In: Ga ratios gathered at 20 mA input

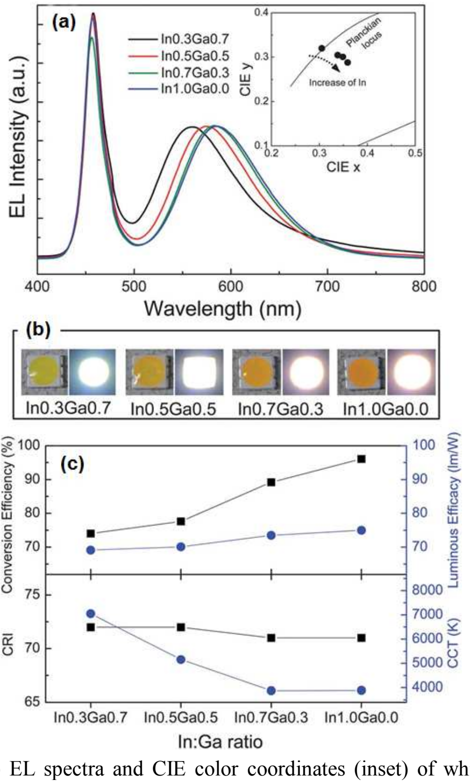

Fig. 8 (a) EL spectra and CIE color coordinates (inset) of white LEDs having CIGS/ZnS QDs with different ratio of In: Ga. (b) Photographs of the respective white LEDs applied current of 20 mA. (c) Variations of conversion efficiency, LE, CRI, and CCT of white LEDs in (b) under the same current. Reprinted with permission from ref. 65. Copyright 2012 Royal Society of Chemistry.

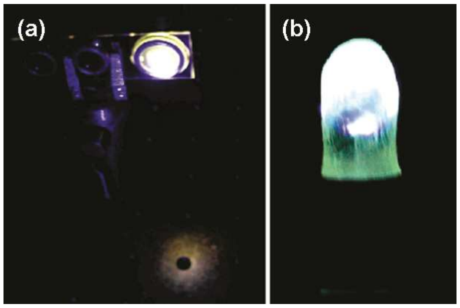

Fig. 9 White-light emission from magic-sized CdSe. (a) Thin film of magic-sized CdSe in polyurethane excited by a frequency doubled titanium: sapphire laser (400 nm) with white light clearly seen reflecting off the table surface. (b) A 5 mm commercial UV LED (400 nm) illuminating a thin coating of magic-sized CdSe in polyurethane. Reprinted with permission from ref. 12. Copyright 2005 American Chemical Society.

current are shown in Fig. 8a. As shown in Fig. 8c, with increasing of In content in the CIGS/ZnS QDs, the conversion efficiency of LED increased from 74 to 96%. Lately, quaternary $ZnCuInS_2$ QDs (ZCIS) and ZCIS/ZnS QDs were also exploited to fabricate white LEDs.[68-69] Despite of these recent progresses, there are still remaining challenges: compared to the binary QDs, the ability to control the size, size distribution, and optical properties of quaternary QDs is still poor.

2.2 White LEDs based on direct white light-emitting QDs

Recently, the development of direct white light-emitting QDs and direct white light-emitting QD-based LEDs are currently a research area of intense interest. Direct white-light generation is photoluminescence of only one type of QD layers that emit white light, which can be excited directly by the light source of UV LEDs.[11] This type of white LEDs can overcome some drawbacks of devices generally fabricated with multiple QDs or phosphor layers, due to self-absorption, scattering, and reflections.[70]

Chen *et al.* demonstrated the fabrication of white LEDs with direct white light-emitting ZnSe QDs, showing great potential for use in lighting applications.[71] In this context, direct white light-emitting ZnSe QDs were synthesized by a colloidal chemical approach using ZnO and Se powder as precursors. The PL of the specimens showed strong white emissions (~200 nm full width at half maximum) in the visible range under ambient conditions. Based on which, the white LEDs were fabricated using a near-UV InGaN chip as the excitation source with CIE chromaticity coordinates of (0.38, 0.41). Also, a CdSe QD-based LED emitting cool white light was successfully obtained by Bowers and co-workers in 2005 (Fig. 9).[12] The as-prepared CdSe QDs (~1.5 nm) exhibited a strong strokes shift and broadband emission (420-710 nm) covering almost the entire visible region. These features above provided a relative balanced white-light emission with CIE chromaticity coordinates of (0.32, 0.37). Subsequently, various methods were attempted in the past to brighten direct white light-emitting CdSe QDs.[70, 72-73] However, in most cases, the efficiency was quite low thereby limiting their further application in white LEDs. To enhance the LE, new modified approaches are highly desired. One improvement by Sapra's group is the independence of the surface-state emission from the QDs, the so-called "emission by chance". They synthesized two different kinds of QDs that were used for white-light illumination, trap-rich CdS with a broad surface-state emission and onionlike CdSe/ZnS/CdSe/ZnS QDs with a dual-color emission, respectively.[74] The PL QYs of them can reach 17% and 30%, respectively. As another instance, Rosenthal's group recently reported that a simple treatment method using formic acid to increase the PL QY of ultrasmall white light-emitting CdSe QDs from 8% to 45%.[75] It should be mentioned that the fabricated LEDs coated with CdSe QDs (QY = 40%) have a 8% higher LE than those previously reported.[76] In addition, doped white light-emitting QD-based white LEDs have also been developed. Tseng and co-workers developed white-light-emitting alloyed $Zn_xCd_{1-x}Se$ QD-based white LEDs illuminated by a commercial UV lamp, wherein a new one-step approach was developed for the synthesis of the QDs stabilized with 3-mercaptopropionic acid (MPA) in the aqueous phase.[77] Sarma *et al.* reported the generation of white light from a Mn^{2+}-doped CdS QDs with an average size of 1.8 nm.[78] By changing the dopant concentration of the as-prepared QDs, different white light could be produced successfully. Most recently, Hickey's group developed a method to produce Mn and Cu co-doped ZnSe QDs (Cu:Mn-ZnSe QDs) *via* a versatile hot-injection colloidal synthesis approach.[25] As a result, the resulted

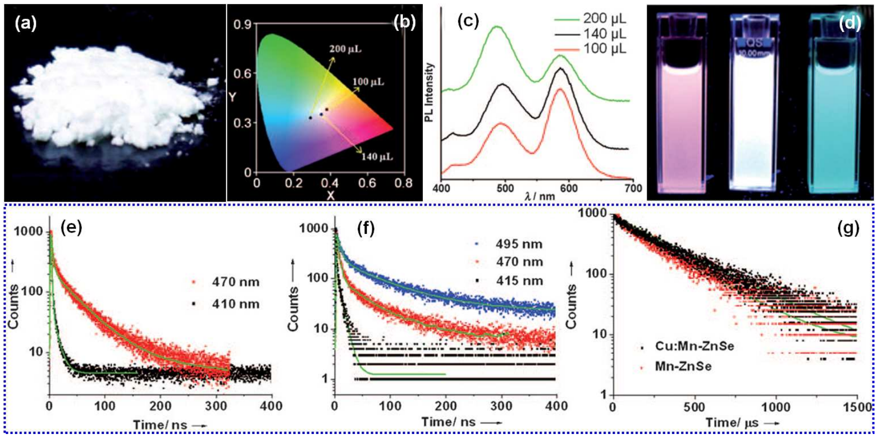

Fig. 10 (a) Cu:Mn-ZnSe doped QD powder showing white-light emission under a UV lamp with a 365 nm excitation. (b) CIE coordinates and (c) emission spectra of QDs with different amount of Cu precursors. (d) Photograph under a UV lamp with a 365 nm excitation. PL decay traces of (e) Mn-ZnSe doped QDs, (f) Cu:Mn-ZnSe doped QDs at different peak positions, and (g) of Cu:Mn-ZnSe and Mn-ZnSe doped QDs at 585 nm. Reproduced with permission from ref. 15. Copyright 2011 Wiley InterScience.

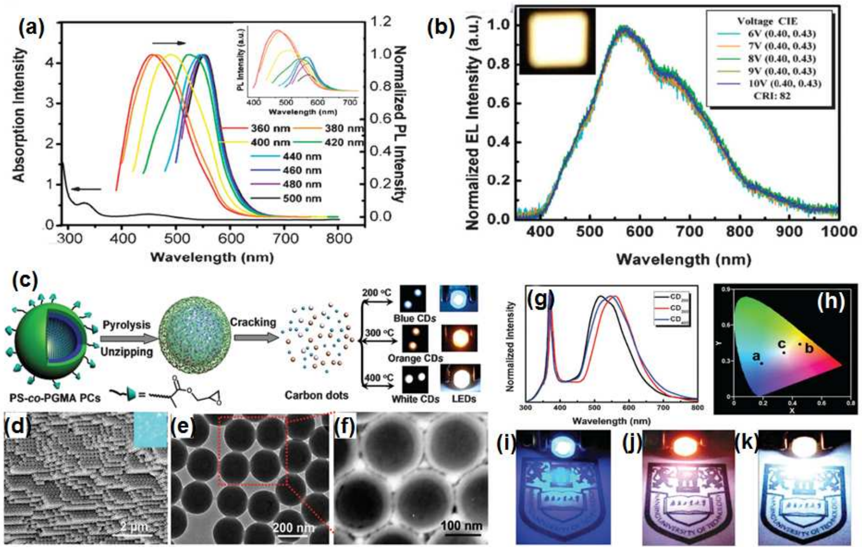

Fig. 11 (a) The absorption and normalized PL spectra of the CDs thin film spin-coated on silica glass. Inset: the normal emission spectra. (b) Normalized EL spectra of as-prepared white LED operated at 9 V. Reproduced with permission from ref. 80. Copyright 2011 Royal Society of Chemistry. (c) Schematic representation of the formation of CDs from pyrolysis of photonic crystals. SEM and TEM images of photonic crystals (d-f). Emission spectra of LEDs (g), CIE chromaticity coordinate (h), and photographs of the corresponding LEDs (i-k), respectively.

QDs possessed high-quality white-light emission in both the colloidal solution and solid-state powder (Fig. 10a) with PL QY as high as 17%. Different emission colors from the doped QDs using varying amounts of the Cu precursor, as demonstrated in Fig. 10b-d. PL decay of the different emission centers in the Mn-ZnSe and Cu:Mn-ZnSe QDs is shown in Fig. 10e-f. Compared to the Mn-ZnSe QDs sample (264 ms), the lifetime of the Cu:Mn-ZnSe QDs could reach up to 324 ms. This kind of doubly doped QDs may hold great promise for the future of white LEDs because it can be synthesized in large-scale with environmentally friendly process. Although the procedure for the fabrication of doped white light-emitting QDs is complex, it offers the possibility of replacement of toxic element-based QD-LEDs.

Another alternative relatively green source of phosphors for white LEDs is heavy metal-free QDs. For instance, a very interesting CZIS QD-based white LED was proposed by Zhang *et al.* in 2011, where the white-light-emitting QDs can be fabricated using commercially available and low toxic precursors (copper acetate, zinc acetate, indium acetate, and sulfur powder).[79] Further, these QDs exhibited tunable PL spectra with emission colors from visible to the NIR region (from 520 nm to 750 nm) and had a relatively high QY (> 70%) without coating any wide band gap shell materials. CZIS QDs prepared by this approach also showed long PL lifetime, large Stoke's shifts, and excellent chemical and thermal stability, in contrast to other colloidal QDs. A simple lighting device was fabricated by using the commercial blue white LEDs covered with CZIS QDs film and the color of the LEDs was transformed from cool blue light into warm yellow light as the chip's emission transmitted through the QD film.

3 Group IV fluorescent nanoparticles based white LEDs

3.1 Carbon-based white LEDs

Carbon-based photoluminescent nanoparticles, including carbon nanodots (C-dots or CDs) and graphene quantum dots (GQDs), are currently a new research topic of intense interest, owing to their favorable optical properties along with their low toxicity, good biocompatibility, photostability, chemical resistance, easy functionalization, and electronic transport properties.[81-86] Such nanomaterials show promising potential in applications such as biomaging, biological labeling, sensing, drug delivery, patterning, coding and optoelectronic devices.[16, 84, 87-88]

CDs are novel fluorescent nanomaterials with sizes below 10 nm, firstly discovered during purification of single-walled carbon nanotubes through electrophoresis in 2004.[89] So far, a variety of strategies have been developed to prepare CDs, including arc discharge,[89] laser ablation/passivation,[90-91] plasma treatment,[85, 92] electrochemical synthesis,[82, 93-96] combustion/thermal/hydrothermal/acidic oxidation,[97-98] supported synthesis,[99-101] solution chemistry methods,[102-104] cage-opening of fullerene,[105] and microwave/ultrasonic methods.[106-109] The obtained CDs usually show excitation-dependent PL feature that the PL peak shifts to longer wavelength as the excitation wavelength increases. However, excited by the UV light source, most CDs present merely blue photoluminescence, thereby restricting the range of their practical applications.

To achieve multiple emission colors of CDs towards applications in LEDs, Wang *et al.* produced photo-stable white-light emitting CDs by surface passivation of crude CDs with poly(ethylene glycol) (PEG_{1500}).[110] In this context, the photoluminescence efficiency of the functionalized CDs can reach up to 10%. Both solution and film of functionalized CDs displayed blue PL (excited at 375 nm) and white PL (excited at 407 nm), respectively. The broad emission band spectrum indicated that the CDs might be suitable for application in white LEDs. However, the PL QY was only about 10%, still lower than those of semiconductor QDs. In our opinion there is a plenty of room for improvement of QYs. Alternatively, Wang *et al*. further developed highly luminescent (PL QY > 50%) CDs *via* a one-step approach in non-coordinating solvent and fabricated the first CD-based white LED (Fig. 11a and b).[80] This work demonstrated

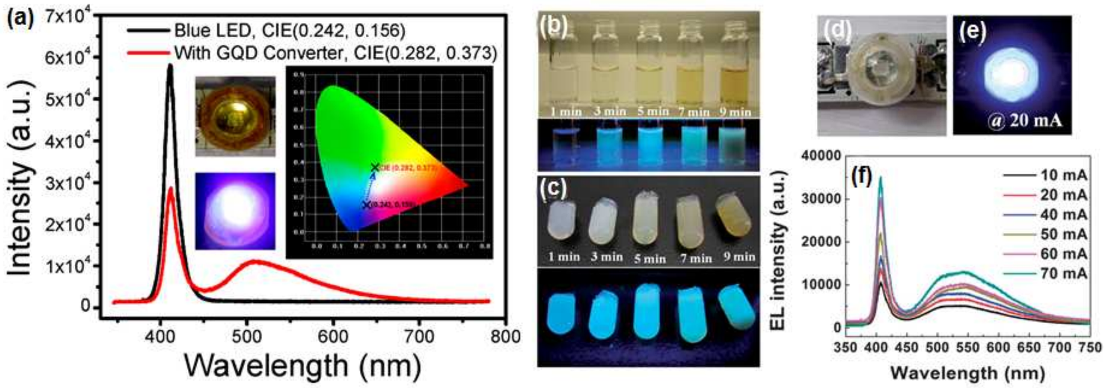

Fig. 12 (a) Color converter of the GQDs. PL spectra of the blue LED with and without GQDs coating. Left inset: Photographs of the GQD coated LED without (*top*) and with applied voltage (*bottom*). Right inset: The CIE chromaticity coordinates for the illuminating blue LED with and without GQD layer. Reproduced with permission from ref. 106. Copyright 2012 American Chemical Society. (b-c) Fluorescence images of the GQD solutions and GQD-agar composites. (d-e) The photograph of the as-fabricated white LED without and with biased current. (f) EL spectra of the white LED under various forward currents. Reproduced with permission from ref. 113. Copyright 2012 Royal Society of Chemistry.

the feasibility of obtaining pure white LEDs with CDs alone, providing an opportunity for simple and high quality light sources with excellent color stability. This opened up new avenues to simple and high quality CD-based light sources that can be compared with natural sun light. Moreover, our group synthesized multicolor fluorescent CDs from chemical unzipping of poly(styrene-*co*-glycidylmethacrylate) (PS-*co*-PGMA) photonic crystals *via* a simple one-step pyrolysis procedure (Fig. 11c-f).[16] And the as-prepared CDs could be well-dispersed in various polar organic solvents and water, and their PL QY could be up to 47% without further surface treatment. Remarkably, they revealed excellent PL properties and good stability, because the powder samples remained unchanged in PL spectra after being stored under ambient conditions for 6 months. With use of the multicolor fluorescent CDs as phosphors, the fabricated LEDs provided bright blue, orange, and warm white illumination. Based on the multicolor fluorescent CDs, the fabricated LEDs provided bright blue, orange, and warm white illumination at (0.19, 0.28), (0.45, 0.44), and (0.34, 0.37), respectively, as displayed in Fig. 10g-k. Besides, another two works of CD-based white LEDs have been also reported recently by our group.[111-112] In one case, a facile plasma-induced approach,[85] was utilized to prepare fluorescent CDs using acrylamide as precursor. These as-prepared CDs were explored as color converters along with CdTe QDs to fabricate white LEDs using a UV-LED chip as the excitation light source. In the other case, a white LED was developed by the combination of the yellow-emitting CDs obtained from the pyrolysis of N-acetylcysteine with blue GaN-based LED chips. Such LED exhibits warm white light with the color coordinates of (0.34, 0.35), very close to the coordinates of balanced white-light emission (0.33, 0.33). These recent advances described above provide new methodologies for the creation of multifunctional CDs, promoting the practical application of CDs in white LEDs.

On the other hand, GQDs and their chemical derivatives have received increasing attention, because of their PL properties as well as their excellent performance of graphene.[114] Their diameters are mainly distributed in a larger range (3-20 nm). Up to now,various top-down and bottom-up methods can be used to synthesize GQDs with tunable size. For instance, highly luminescence GQDs with an average diameter as small as 1.65 nm was prepared by a microwave-assisted hydrothermal method.[106] The QYs of the as-prepared GQDs were determined to be 7-11%. As indicated in Fig. 12a (*left inset*), the results demonstrated that the emission wavelength of the GQDs was independent of the size of the GQDs, which is different from the CDs prepared by conventional methods. Subsequently, a white LED with CIE chromaticity coordinates of (0.28, 0.37) was constructed with GQDs acting as an efficient light converter (Fig. 12a, *right inset*), opening up new avenues for developing deep ultraviolet photonic devices using GQDs as the active material. Luk *et al.* recently demonstrated a microwave-assisted pyrolysis procedure to fabricate fluorescent GQD and GQD-polymer composites by using polysaccharides such as agar (Fig. 12b and c).[113] Moreover, a white LED was demonstrated by coating this GQD-agar composite onto a blue chip (Fig. 12d-f). The GQD-polymer based white LED exhibited over 61% light-conversion efficiency, and displayed better colour stability for over 100 hours. Despite some remained drawbacks, these carbon-based white LEDs should receive more attention due to their low toxicity. In order to promote the industrialization of carbon-based white LEDs, much work is still highly needed in this area, such as utilizing alternative starting materials, improving QYs of CDs, and finding effective strategy in practical applications.

3.2 Silicon QD-based white LEDs

As a basic material of semiconductor and electronic fields, silicon is widely used in chemical industries, integrated circuits and semiconductor devices. However, bulk Si has an indirect gap, which makes it inadaptable for use in many optical-related fields. Recently, luminescent silicon QDs (Si QDs) are highly attractive for LEDs because they are considered to be nontoxic, photostabe, conductive, and environmental-friendly nanomanufacturing.[115-117] Towards this end, exploitation of light emission from nanostructured Si and the improvement of the optical properties are very important.

Ever since Canham firstly demonstrated luminescent Si nanocrystallites derived from anodic electrochemical etching of

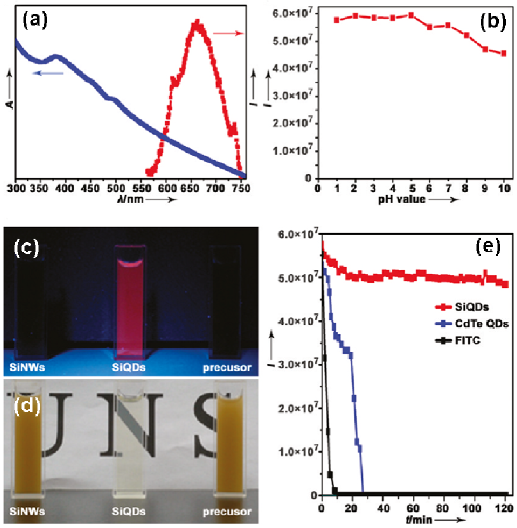

Fig. 13 (a) UV-PL spectra of the as-prepared Si QDs. (b) Temporal evolution of fluorescence of the Si QDs under various pH values. Photographs of aqueous solutions dispersed with the free-standing Si nanowires (*left*), as-prepared Si QDs (*middle*), and reaction precursors (*right*) under 365 nm irradiation (c) and ambient light (d). (e) Photostability comparison of as-prepared Si QDs. Reproduced with permission from ref. 128. Copyright 2011 American Chemical Society.

bulk Si,[118] quantum-confined light-emitting silicon-based nanomaterials have drawn considerable research.[119-121] And a variety of approaches have been exploited to fabricate Si QDs, including electrochemical etching,[122-126] thermal vaporization,[127] pyrolysis (e.g., laser heating, microwave, microdischarge and plasma treatment),[129-136] and wet chemistry approaches.[137-139] One challenge in the synthesis of Si QDs is to overcome their size polydispersity. To obtain monodispersed Si QDs, a new one-pot microwave-assisted method was demonstrated to synthesize water-dispersible fluorescent Si QDs using Si nanowires and glutaric acid as precursors (Fig. 13).[128] Significantly, the as-prepared Si QDs featured excellent aqueous dispersibility, which made it suitable for optoelectronic applications such as LEDs. The as-prepared Si QDs possessed good optical properties with clearly resolved absorption peak and symmetrical PL peak (maximum emission wavelength at ~660 nm) (Fig.13a). It clearly shows that the as-prepared Si QDs feature excellent aqueous dispersibility (Fig. 13c and d), which is attributed to the large amount of surface-covered glutaric acid with hydrophilic carboxylic groups. Based on microwave-assisted method, 3-aminopropenyl-terminated Si QDs exhibiting intrinsic PL QY of 15% were also successfully prepared.[140]

Another straightforward method, the so-called density gradient ultracentrifugation, was introduced for obtaining the first examples of monodisperse fractions of Si QDs.[141] Density gradient ultracentrifugation is a size-separation technique that has been researched extensively for purification of single-walled carbon nanotubes (SWNTs) with both aqueous and organic density gradient media.[142-143] The monodisperse Si QDs were prepared by size-separation of polydisperse alkyl-capped Si QDs, that were synthesized by thermal processing of trichlorosilane-derived sol-gel glasses followed by HF etching and surface passivation with alkyl chains and were subsequently fractionated by size using a self-generating density gradient of 40 wt % 2,4,6-tribromotoluene in chlorobenzene. As a result, density gradient ultracentrifugation turned out to be a useful technique for the quantification of Si QDs with size-dependent optical properties [144-145] In addition, wavelength-controlled etching,[146] hydrosilylation of layered polysilane (Si_6H_6),[147] sonochemical route,[117] template-directed self-assembly,[148] *etc.* are also demonstrated to prepare high-performance fluorescent Si QDs. For another instance, a direct white light emitting Si QDs (Fig. 14b) were prepared at room temperature and ambient pressure *via* sonochemical route, and the luminescence of the as- obtained Si QDs (about 2.8 nm) was observed in the wide range between 340 nm and 700 nm (Fig. 14a).[117]

Based on luminescent Si QDs as phosphors, various LEDs were fabricated. Cheng *et al.* demonstrated a Si QD-based LED, though there were some disadvantages in terms of poor spectral purity.[149] To optimize their performance, Puzzo *et al.* described tunable visible electroluminescence from a hybrid Si QD-based LED.[116] However, the EQEs of the as-prepared device is only about 0.7%, significantly lower than those obtained using II-VI counterparts. It is noteworthy that most recently an optimized Si QD-based LED with highly efficient electroluminescence was successfully fabricated with the EQEs could catch up to 8.6%.[150] To further enhance device efficiencies, unremitting work is still underway.

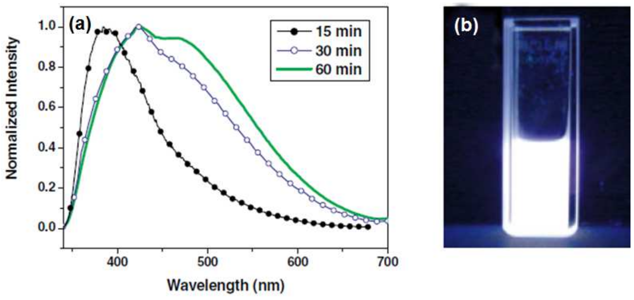

Fig. 14 (a) Room temperature PL spectra of Si QDs excited with 325 nm He-Cd laser. (b) The photograph of as-prepared Si QD sample under the UV lamp (360 nm). Reproduced with permission from ref. 117. Copyright 2004 Wiley InterScience.

Si QD-based LEDs represent a good prospect application in solid-state lighting in the near future, due to their low toxicity and stability. To date, there are still some problems urgent to be solved. On one hand, the QYs of Si QDs is too low, thereby limiting their further application. On the other hand, the process of fabrication is complicated. Moreover, up to now less species of Si precursor have been found. And more novel start materials and effective strategies are still needed to exploit.

4 White LEDs based on organic-inorganic fluorescent nanocomposites

Inorganic-organic fluorescent nanocomposites combine the unique properties of inorganic quantum-confined nanoparticles with polymers possessing excellent stability, fluorescence, flexibility, processability and mechanical properties.[151-153] These nanocomposites are novel building blocks and hold promise for use in photovoltaic devices. To date, a variety of approaches have been devoted to designing and constructing novel organic-inorganic fluorescent nanocomposites.[13, 72, 154-160]

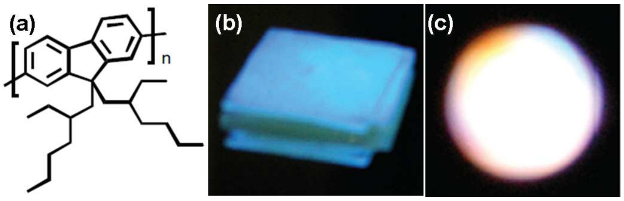

Fig. 15 (a) Chemical structure of 9,9-bis(2-ethylhexyl)polyfluorene and (b) photograph of its PL in blue. (c) Images of white light emission from hybrid QD-conjugated polymer based white LED: yellow, green and red QDs (PL = 580, 540 and 620 nm) and blue polyfluorene (PL = 439 nm) hybridized on a UV LED (EL = 383 nm). Reproduced with permission from ref. 13. Copyright 2007 IOP Publishing.

4.1 White LEDs based on layers of QDs and fluorescent organic conjugated polymers

White light generation employing different polymers has previously been studied.[161] In a variety of polymer types, fluorescent organic conjugated polymers are specifically promising candidates in solid-state lighting applications in the near future. On the one hand, they have very strong absorption in near-UV on the order of 10^5 cm^{-1} with high EQE. On the other hand, polymers can be deposited easily with ordinary techniques such as spin coating. Take advantages of these significant performances, Kim and co-workers fabricated white LEDs based on UV light sources with conjugated polymers-QDs as converters.[162] In this context, they combined the 380-nm UV LEDs with the composite of blue-emitting conjugated polymer (poly[(9,9-dihexylfluorenyl-2,7-diyl)-alt-*co*-(2-methoxy-5-{2-ethylhexyloxy}-1,4-phenylene)], BEP), yellow green-emitting copolymer (poly[(9,9-dioctylfluorenyl-2,7-diyl)-*co*-(1,4-benzo-{2,1', 3}-thiadiazole)], GEP), and 605-nm CdSe/ZnS QDs as hybrid phosphors for fabricating white LEDs with high CRI. As a result, high-powered device were obtained with CIE coordinate of (0.38, 0.37), CRI of 90.0, and CCT of 3969 K, respectively, at 20 mA.

Heliotis *et al*. developed microscale white light sources by the combination of conjugated polymers and UV micro-LED arrays.[163] The as-prepared devices were based on blue-, green- and red light-emitting polyfluorene materials converting the emission from an array of UV micro-LEDs. This provided an appealing route for us to develop microscale white light sources, which can applicate in microdisplays. Additionally, Demir *et al*. demonstrated a white light generation by dual hybridization of QDs and conjugated polymers (Fig. 15a and b) for the first time.[13] As shown in Fig. 15c, the generated white light was adjusted by layer-by-layer assembly of CdSe/ZnS QDs and polyfluorene conjugated polymer deposited on near-UV LEDs. By utilizing such layer-by-layer hybridization, individual QDs film thicknesses were well controlled and adjusted precisely.

4.2 White LEDs based on QDs nanocomposites with functionalized polymer derivatives

A series of diverse hierarchical structures (wire-like, belt-like, and even sheet-like structures) of homogeneous QDs with highly photoluminescent properties have been proposed by our group *via* the molecular-assembly procedure.[164-167] Very recently, catalytic chain transfer polymerization (CCTP) technique was utilized to fabricate poly(methacrylic acid) (PMAA) macromonomers ligands (Fig. 16a). Based on the as-synthesized macromonomers ligands, our group successfully prepared CdTe QDs nanocomposites as color convert materials towards warm white LED devices (Fig. 16b and c).[168] The as-prepared LED device exhibited the CIE coordinates of (0.35, 0.36) and the spectrum of which is shown in Fig. 16d and e. It is believed that the reported route offers an available pathway to achieve robust QDs for white LEDs applications. Additionally, most recently our group also reported the facile fabrication of white light-emitting CdS-poly(2-hydroxyethyl acrylate (HEA)-*co*-N-vinylcarbazole (NVK)) nanocomposites from the incorporation of 3-(trimethoxysilyl)-1-propanethiol (MPS)-capped CdS into carbazole-containing polymer *via* plasma-ignited frontal polymerization (PIFP).[169] The schematic illustration of the formation of CdS-poly(HEA-*co*-NVK) fluorescent nanocomposites prepared by PIFP is demonstrated in Fig. 16f. As shown in Fig. 15g, poly(HEA-*co*-

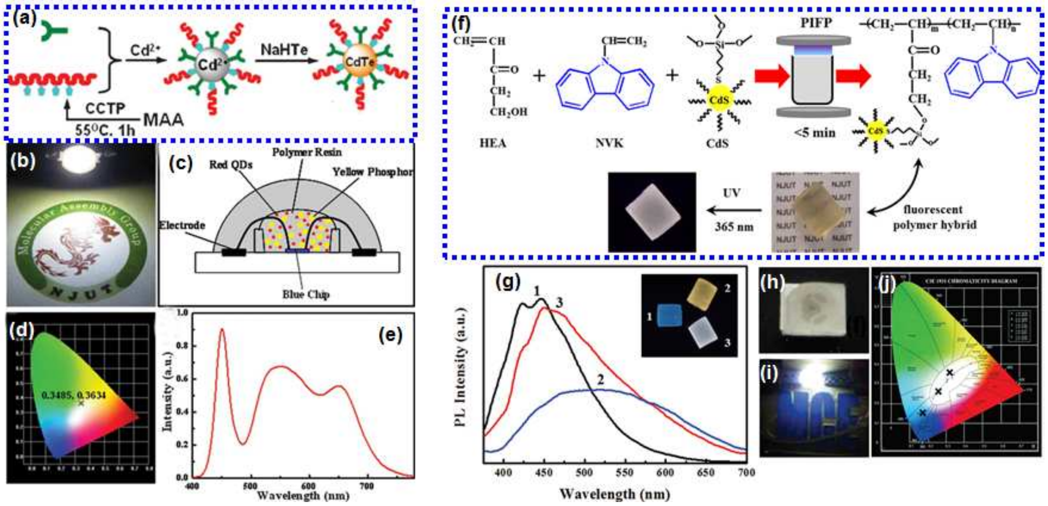

Fig. 16 (a) Illustration of the route for the synthesis of CdTe QDs. (b) Photograph of the as-prepared LED device in the dark. (c) Schematic view of the fabrication of the white LED device with QD phosphor nanocomposites. The CIE color coordinates (d) and emission spectrum (e) of the white LED. Reproduced with permission from ref. 168. Copyright 2012 Royal Society of Chemistry. (f) Schematic illustration of the preparation of fluorescent nanocomposites. (g) PL emission spectra of poly(HEA-*co*-NVK) and CdS nanocomposites. (h-i) Digital pictures of the as-fabricated white LED without and with biased current. (j) CIE diagram of the corresponding QD-LED. Reproduced with permission from ref. 169. Copyright 2012 Wiley InterScience.

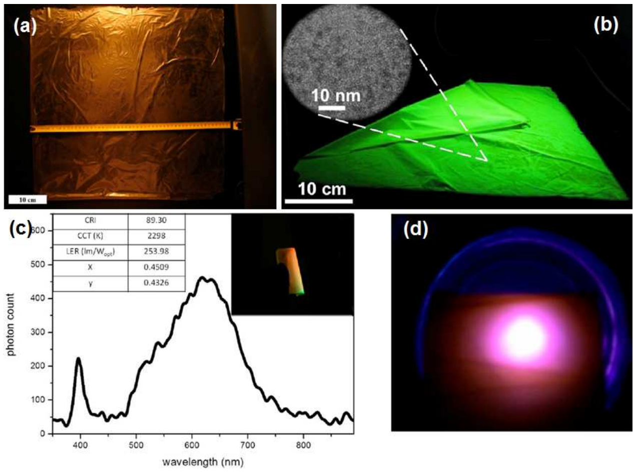

Fig. 17 Photograph of a 51 cm × 51 cm InP/ZnS QD hybrid film under room light along with a ruler (a) and the folded film under UV illumination (b). (c) EL spectra of a proof-of-concept white LED using an InP/ZnS QD film as the remote color-converting nanophosphors. (d) Photograph of an exemplary device under operation. Reproduced with permission from ref. 171. Copyright 2012 American Chemical Society.

NVK) presents an emission wavelength at about 450 nm, showing blue light upon UV excitation, whereas CdS-PHEA exhibits the emission wavelength at about 540 nm, presenting yellow emission upon UV excitation. White LEDs were fabricated with its emission wavelength spreading the visible range from 400 to 700 nm, by facile integration of the broad yellow spectrum with its compensating color blue light. Moreover, the chroma of white emission can be easily tuned from blue at CIE coordinates of (0.17, 0.16) to warm white (0.31, 0.36) by adjusting the weight ratios of CdS/NVK (Fig. 16h-j). This simple and energy-saving method provided an available pathway to manufacture white LEDs based on fluorescent nanocomposites. In another study, ionomer-derived honeycomb-patterned PMMA/ZnS QDs films with favourable fluorescence, optical performance and hydrophobicity, were fabricated by our group *via* a breath figure (BF) method.[170] As a result, the LEDs coated with these as-prepared honeycomb architectures show 10.3% and 17.7% improvement of LE, which suggests that the multifunctional films are valuable for the fabrication of LEDs.

4.3 White LEDs based on QD/CD-polymer hybrid film

To construct optoelectronic devices, flexible and large-scale photonic materials especially in terms of stand-alone flexible films are strongly required. For this purpose, flexible and freestanding InP/ZnS QDs films have been developed as remote color-converting nanophosphors for white LED applications, as shown in Fig. 17.[171] The flexible QD-polymer films were successfully fabricated over very large areas of greater than 50 × 50 cm (Fig. 17a and b). The TEM images demonstrated the uniformity of the QDs dispersed within the host PMMA. The QD nanocomposite films allow for high CRI, warm white light generation with CRI of 89.3 and CCT of 2298 K (Fig. 17c and d). It has been proved that polymer-based nanocomposites open up new possibilities for innovative eco-friendly LEDs devices by embedding semiconducting QDs synthesized by colloidal chemistry. In a similar work, Kwon fabricated large-scale (20 × 20 cm) independent luminescent films based on the CDs, which were made from polyacrylamide (PAA) *via* emulsion-templated carbonization method.[17] By combining the CDs film and blue chip (400 nm) they demonstrated a white LED, and the LE of which could reach as high as 108.2 lm Wopt^{-1}, almost identical to QDs or compound phosphors. The results showed that hybrid luminescent films provided a promising remote fluorescent materials for future white LEDs.

5 Summary and outlook

In this feture article, we have highlighted the recent advances in white LEDs constructed from different fluorescent nanomaterials, including nanocrystal quantum dots, carbon nanoparticles, graphene quantum dots, silicon quantum dots and organic-inorganic fluorescent nanocomposites. The development of fluorescent nanomaterials opens novel avenues for the device of light source of white LEDs. In contrast to conventional fluorescent materials such as inorganic phosphors, the white LEDs based on fluorescent nanomaterials display superior optical performance similar to sunlight, due to their unique and tunable optical properties. Among them, carbon-based white LEDs and Si QD-based white LEDs have drawn more attention because of their low toxicity and stability compared with QD-based white LEDs. In particular, inorganic-organic fluorescent nanocomposites provide better flexibility, processability, stability and mechanical properties for white LEDs, largely expanding their practical applications. And the fluorescent nanomaterial-derived white LEDs will be rapidly developed in the near future.

Although fluorescent nanomaterial-derived white LEDs have displayed enormously exciting potential applications, some vital hinders also exist before their commercialization. The following key issues should receive greater attention in the future. Firstly, the photostability and thermo stability of the fluorescent nanomaterials are not as good as conventional phosphors, and thereby further researches on this repsect are highly needed. Secondly, the productivity of fluorescent nanomaterials is very low, which limits their large-scale application. Thereby, it is important to develop large-scale films and powders in the future. Another remarkable drawback is the complex process of fabrication. And accordingly, facile and effective techniques are desired to exploit. Finally, how to reduce the cost is a key issue. Nevertheless, with the improvement in techniques, we strongly believe that new powerful possibilities in the field of fluorescent nanomaterial-derived white LEDs will be explored.

Acknowledgements

This work was supported by the National High Technology Research and Development Program of China (863 Program) (2012AA030313), National Natural Science Foundation of China (21076103), Specialized Research Fund for the Doctoral Program of Higher Education of China (20103221110001), Industrial Project in the Science and Technology Pillar Program of Jiangsu Province (BE2012181), and Priority Academic Program Development of Jiangsu Higher Education Institutions (PAPD) and the Colleges and Universities in Jiangsu Province Plans to Graduate Research and Innovation (Grant CXZZ12_0449).

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