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Nanomedicine & Nanotechnology Open Access
ISSN: 2574-187X

Recent Advances of Carbon Dot-Based Light-Emitting Diodes

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Mini Review

Volume 2 Issue 2

Received Date: March 21, 2017

Published Date: March 23, 2017

DOI: 10.23880/nnoa-16000120

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Mini Review

Carbon dots (CDs), with abundant raw materials on earth, have become a new-type of luminescent materials and received great attention for their excellent merits such as low cost, low toxicity, strong photo stability, good biocompatibility, and high photoluminescence (PL) [1-3]. It's superior to traditional fluorescent organic dyes and luminescent inorganic quantum dots (QDs).

Synthetic approaches for CDs can be classified into two general routes: top-down and bottom-up methods. The former refers to the cutting from different carbon sources into CDs. The methods include laser ablation [4], chemical ablation [5], electrochemistry [6], arc discharge [7], microwave irradiation [8]. The latter be realized by dehydration and carbonization of small molecules or polymers. Bottom-up methods involve hydrothermal or solvothermal treatment [1,9], pyrolysis [10], template [11] and microwave assisted synthesis [12], chemical oxidation [13].

The optical properties of CDs are mostly studied, because they are important for investigating the PL mechanism and adapting CDs for various applications. In general, the emission peaks of CDs are almost symmetrical on the whole wavelength scale. The fluorescence spectra are usually wide and exhibit large Stokes shifts compared with semiconductor QDs and organic dyes. The PL of CDs usually shows excitation-dependent behavior, which may result from the wide distribution of the size, surface defect states or different emissive traps [14]. Fortunately, the excitation-dependent PL behaviors make CDs promising for multicolor imaging applications [10,15]. Although many efforts have been made, the PL mechanism of CDs is still under debate. Two popular PL mechanisms have been mostly discussed: the bandgap transitions in the presence of sp^2 clusters where the quantum confinement effect features prominently [9,16], and the surface defect states [6,17].

After years of research, blue fluorescent CDs in aqueous solution with a quantum yield (QY) higher than 80% can successfully prepared, which is even comparable with the best inorganic semiconductor QDs [2]. Green emissive CDs with PL QY of 55%were realized after fractionating on an aqueous gel column and surface passivation [18]. Orange luminescent CDs with PL QY of 46% were fabricated through surface charge engineering by surface metal-cation fictionalization [9]. These achievements make CDs promising candidates for the application of LEDs.

Nowadays, LEDs have drawn worldwide attention for their potential applications in liquid-crystal displays as white backlight and full-color displays as well as the next generation of lighting sources [19,20]. CDs can be expected to substitute for phosphors in LEDs for their low cost, low toxicity and tunable stable fluorescence emission compared to rare-earth-based phosphors or traditional toxic metal-based semiconductor QDs. Consequently, some efforts have been made to fulfill the applications of CDs in LEDs. CDs can work both as phosphors component in color-conversion LEDs or active layers in electroluminescent (EL) devices.

The phosphor based LEDs usually consist of UV or blue chips and fluorescent CDs. One of the problems with the CDs as phosphor is the weak fluorescence due to aggregation induced quenching. To avoid it, many groups have embedded CDs in solid matrices such as silicone [21], starch [22], resin [23], PMMA [24] and PVP [25], to preserve the fluorescence. Recently, Wang et al. fabricated liquid LEDs and solid LEDs based on the CDs with excitation wavelength independent but solvent-dependent emissions [26]. The liquid-form devices with nice emission colors of green, yellow and red were prepared through sealing the CD solution in a flat silica glass box and packing above a UV-LED chip (Figure 1a–c). CDs in liquid format can solve the aggregation induced quenching effect in solid state [15]. The CDs were also mixed with polymers and dropped on a UV-LED chip for the preparation of solid LEDs (Figure 1d–f). This is the first report that the same CDs emit different colors when mixed with different polymers under one single excitation wavelength. These LEDs were based on the same CD particles and demonstrated promising application potentials. However, the CD-LEDs are still with relatively low efficiencies compared with the well-developed QD-LEDs. Therefore, new synthetic methods for large-scale production of CDs with high QYs need to be developed.

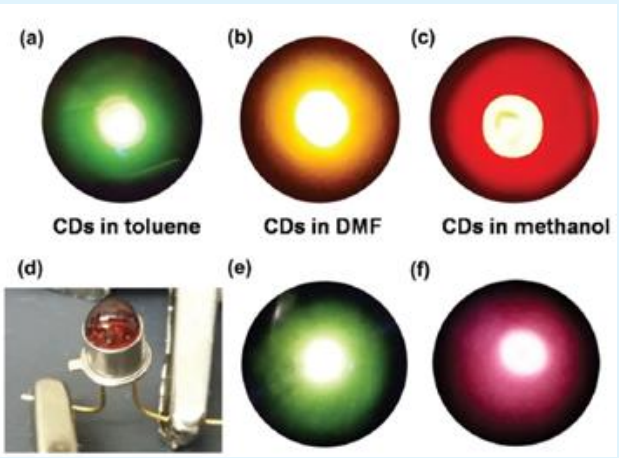

Figure 1: The photographs of CD-LEDs: emissions of liquid LEDs in toluene (a), DMF (b), and methanol (c); solid LEDs of CD/polymer under room light (d) and the emissions of CD/PMMA (e) and CD/PVA (f). Adapted from reference [26].

For EL devices, Zhang et al. reported LEDs with driving current controlled color of blue, cyan, magenta, and white based on highly blue fluorescent CDs [27] (Figure 2). The devices consisted of a carbon dot emissive layer sandwiched between an organic whole transport layer and an organic or inorganic electron transport layer fabricated by a solution-based process. The interesting current density-dependent emission is useful for the development of multicolor LEDs. Yuan et al. fabricated the bandgap fluorescent CDs from blue to red with QY up to 75% for blue fluorescence which was achieved by the solvothermal method [28]. The CDs were the active emission layer for monochrome electroluminescent LEDs, and the maximum luminance reached 136 cd m^{-2} for blue LEDs. The LEDs all show substantial stable EL and voltage-independent emission color, which is significant for display and lighting technology. Moreover, white LEDs with CIE coordinate of (0.30, 0.33) and a luminance of 2050 cd m^{-2} were also fabricated. Nonetheless, the research of CD-based EL LEDs is still at their very early stage. There are two main challenges: the much higher QY of CDs in the solid and thin film states, as well as the optimization of electron and whole transport property of CDs.

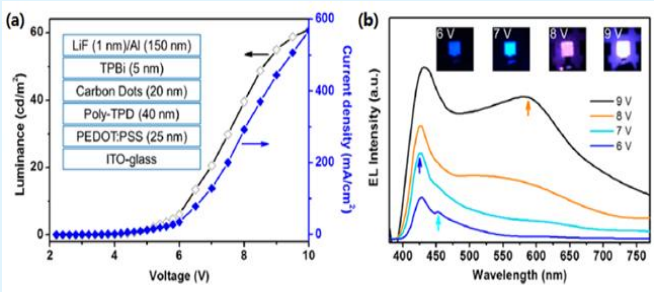

Figure 2: (a) Current density and brightness of the CD-LEDs, with an inset showing the device structure; (b) EL spectra and true color photographs of blue, cyan, magenta, and white emissions. Adapted from reference [27].

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