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Improved performance of flexible white hybrid light emitting diodes by adjusting quantum dots distribution in polymer matrix



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ABSTRACT

A flexible hybrid white light emitting diode (LED), in which the CdSe/ZnS quantum dots (QDs) acted as an orange emitter and small organic molecules as the blue emitter in a multilayered structure was fabricated by spin-coating. The poly (N-vinylcarbazole) (PVK) was added in the emitting layer to adjust QDs distribution in polymer matrix. By increasing the concentration of PVK, the electroluminescent intensity at 610 nm emission was enhanced since QDs had a better dispersibility in polymer matrix with the presence of PVK, and a pure white light emission was obtained due to the optimized architecture of the hybrid active layer. Our study provides a novel method for improving the device performance of hybrid white LEDs by controlling the QDs distribution in the active layer.

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Since the first announcement of quantum dot light emitting device (QD-LED), an extensive research has been established because of its advantages such as low power consumption, long lifetime, large area lighting, low driving voltage, flexibility and the easy fabrication process using variety of QDs which can be deposited by spin casting, inkjet printing, and dip coating [1–8]. Up to now, white colour devices are the most attractive emitting colour because of their benefits in displays back lighting and large area displays [9–11]. Therefore, it is important to develop white light emitting devices based on QD materials.

So far, a few strategies have been proposed to achieve white QD-LEDs [12–14], for instance, a prototype results from the direct electroluminescence (EL) of QDs through integrating red, green, and blue emitting QDs to produce high-efficiency white devices [15,16]. However, the synthesis of high quantum yield of deep-blue-emitting QDs is rare with low efficiency [17]. Therefore, one alternate approach is to combine multiple emissions from different recombination regions in organic/inorganic hybrid optoelectronic structures, which can be used to obtain white light emissions [18,19]. Recently, Zhang et al. [20] reported the fabrication of white-emitting devices containing a bilayer QDs/polymer structure in the device active region. Besides, Chou et al. [21] reported an organic/

inorganic hybrid white-emitting light emitting device by integrating core-shell CdSe/ZnS quantum dots as a yellow emitter and polyfluorenes as the blue emitter in a multilayered structure. However, these devices encountered a problem that the QDs had a poor dispersibility in polymer matrix.

Herein, in this work, we report the fabrication and characterizations of a hybrid white LED essentially based on a structure formed by blue emitting small molecules 4, 4-bis (2, 2diphenylethen-1-yl) biphenyl (DPVBi) and orange emitting CdSe/ ZnS QDs. In order to adjust quantum dots distribution in polymer matrix, the poly (N-vinylcarbazole) (PVK) was added into the mixture solution of DPVBi and QDs. The performance of these devices with the different concentrations of PVK was investigated in detail.

The flexible polyethylene terephthalate (PET) was used as the substrate. The indium-tin-oxide (ITO)/PET substrate was purchased from Csgholding in Shenzhen, China. The sheet resistance was 150 Ω /sq. The CdSe/ZnS QDs was obtained from Ocean Nanotech in America with an emission peak at wavelength of 610 nm, and a full-width at half-maximum (FWHM) of 25 nm. The poly(N,N'-bis- (4-butylphenyl)-N,N'-bis(phenyl)benzidine) (poly-TPD) was purchased from Lumtec in Taiwan. All the materials, lithium fluoride (LiF), tris-(8-hydroxyquinoline) aluminium (Alq₃), 4,7-diphenyl-1,10-phenanthroline (Bphen), DPVBi and PVK were purchased from Aglaia in Beijing, China.

Fig. 1(a) shows the schematic of the as-fabricated devices on flexible ITO/PET substrate. As shown in Fig. 1(a), the flexible white



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Fig. 1. (a) Device configuration of the flexible hybrid white LEDs based on small molecules and quantum dots. (b) Chemical structures of the organic materials used in this work.

hybrid LEDs have a device structure of PET/ITO/PEDOT:PSS/poly-TPD/PVK:DPVBi:QDs(x:2:1)/Bphen/DPVBi/Bphen/Alq₃/LiF/Al. The chemical structures of the organic materials used in this work are shown in Fig. 1(b). The ITO/PET substrates were cleaned for 20 min by ultrasonic cleaning in acetone, and absolute ethyl alcohol, respectively. Then, the ITO/PET substrates were subjected to 5 min plasma treatment for further cleaning. The buffer layer consisting of conducting polymer polv(3.4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS) and 2% concentration of dimethyl sulfoxide (DMSO) was deposited on ITO/ PET substrates by spin coating at 5000 rpm for 30 s, and dried at 150 °C for 10 min. The purpose of the introduction of DMSO was to enhance the conductivity of the polymer layer. Subsequently, a poly-TPD layer and a PVK:DPVBi:CdSe/ZnS QDs hybrid layer were formed by using spin-coating technique with a speed of 2000 rpm for 30 s.

For the preparation of the hybrid layer, the PVK:DPVBi:CdSe/ZnS QDs mixture in toluene was prepared by mixing 10 ul of PVK (with varying concentrations, *x* mg/mL), 40 ul of CdSe/ZnS (4 mg/mL) and

80 ul DPVBi (15 mg/mL) under 10 min vigorous stirring. The concentration of PVK was varied from 0 mg/mL to 20 mg/mL. After that, the Bphen(10 nm)/DPVBi(5 nm)/Bphen(10 nm)/Alq₃(5 nm) layers were thermally evaporated at 3×10^{-4} Pa on the PVK:DPVBi:CdSe/ ZnS QDs hybrid layer. Finally, 0.4 nm LiF and 100 nm Al were thermally deposited on the samples at 2×10^{-3} Pa to form the metal contacts. The active area of the cells was 5×10 mm².

The current density–voltage (J-V) measurements were performed with Fluorolog-3 instrument. The electroluminescence (EL) spectra and photoluminescence (PL) spectra were recorded with a Hitachi F-4600 fluorescence spectrophotometer. The luminous intensity and Commission International De l'Eclairage (CIE) coordinates were measured using a Topcon SR-3A spectroradiometer. All tests were carried out at room temperature in atmospheric pressure.

Fig. 2(a) shows the corresponding EL spectra of the devices with varied concentrations of PVK at the bias voltage of 15 V. For all the concentrations, the EL spectra exhibits two primary peaks at 460 nm and 610 nm, and a small shoulder peak at around 490 nm. It



Fig. 2. (a) EL spectra of the devices with varied concentration of the PVK (0 mg/mL, 5 mg/mL, 10 mg/mL, and 20 mg/mL, respectively). (b) PL spectra of Alq₃, poly-TPD, CdSe/ZnS QDs, DPVBi and Bphen. (c) Energy level diagram of the flexible hybrid white LEDs. (d) PL spectra of the hybrid film of DPVBi:poly-TPD with various weight ratios.

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