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Efficient and angle-stable white top-emitting organic light emitting devices with patterned quantum dots down-conversion films

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ABSTRACT

Efficient white top-emitting organic light-emitting devices (TEOLEDs) are fabricated by combining blue TEOLEDs with down-conversion films. Quantum dots (QDs) are integrated with polymethyl methacrylate (PMMA) to obtain freestanding composite films, and the films are used as down-conversion films to produce red emission by converting blue emission from blue TEOLEDs. The blue and white TEOLEDs exhibit the maximum current efficiency of 37.4 and 22.2 cd/A, respectively. Through integrating a microstructure QDs down-conversion film by transfer printing on blue TEOLED, the light extraction performance of white TEOLEDs is improved and the angular distribution of the white emission is almost Lambertian. The CIE color coordinates of white TEOLEDs with patterned QDs down-conversion films shift only (0.025, 0.005) from 0° to 60°.

1. Introduction

Compared with the conventional white bottom-emitting OLEDs, white top-emitting OLEDs (TEOLEDs) are preferred because a higher aperture ratio allows us the facile choices of substrates, including opaque ones. So, they have unique advantages in the fields of display and lighting [1]. But it is difficult to fabricate high-quality white TEOLEDs. Because the strong microcavity effect introduced by the semitransparent top electrode can lead to the strong dependence of color coordinates on the viewing angle [2–4]. So, there are many extensive efforts focused on eliminating the microcavity effects to obtain white TEOLEDs [5–7]. The combination of ultrathin metal electrode with light extraction layer of high refractive index can effectively eliminate the microcavity effect and improve the angular stability [8]. However, the small thickness of ultrathin metal electrode reduces the film density [9], which brings the poor stability and reduces the lifetime. Besides, white devices require combining at least two complementary colors. One traditional way to fabricate white OLEDs is stacking two or more different emitting layers in the emissive region. Its disadvantages are the complex device structure and the shifting of recombination region as driving voltages increasing [10]. It means that the color will change with different voltages. And they also require careful fabrication process in order to maintain a high production yield [11]. Another way to fabricate white TEOLEDs is mixing different organic electroluminescence (EL) materials in a single emitting layer. However, the degradation rates of different organic EL materials are different, resulting in the distortion in color over time [12].

Down conversion structure is an important way to achieve white TEOLEDs [13–15]. We first achieved white TEOLEDs based on down conversion structure in 2008 [13]. The blue light pumped by TEOLED excites the down-conversion materials, and white light is achievable through a complementary bicolor mixing of blue-pumping light from the TEOLED and the light converted by the down-conversion materials. The color stability against voltage and operation time can be easily achieved on account of the synchronous changes of the different colors. But there are still some issues that remain to be resolved. On the one hand, the absorption spectra of the organic down-conversion materials are in the ultraviolet (UV) region, so the principal deficiency is less utilization of the blue light which resulting in low efficiency. On the other hand, the viewing angle dependency caused by the microcavity needs to be improved.

Quantum dots (QDs) are employed as perfect down-conversion materials due to the size-dependent emission wavelength and high photoluminescence (PL) quantum yield [16–20]. The beneficially high absorption capability at wavelengths of around 500 nm can provide high conversion efficiency. As inorganic material, the lifetime of QDs is longer than that of organic materials. Unlike organic small molecules, it is not possible to deposit QDs by thermal evaporation owing to their high molecular weight [21]. In general, QDs are dissolved in an organic or inorganic solvent. And excellent miscibility of QDs with polymer in polar solvent enables the uniform dispersion of QDs within the polymer host. Hence, it is feasible to stabilize the QD solution by converting it

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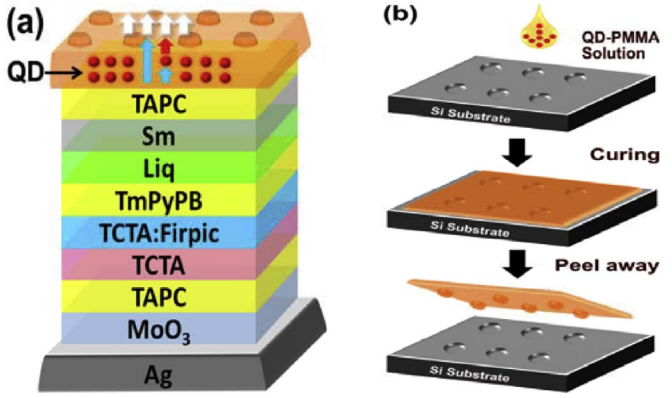

Fig. 1. (a) Device structure of the white TEOLED combining BTEOLED with patterned down-conversion film. (b) Schematic process flow for the patterned down-conversion films.

into a thin film [22,23]. Thus, efficient white LEDs or OLEDs based on QDs down-conversion films were reported [24–29]. However, to our knowledge, there is no report on white TEOLED with QDs down-conversion films.

In this work, white TEOLEDs are demonstrated by combining blue TEOLEDs with QDs down-conversion films. The blue TEOLEDs with very high efficiency are used as pumping sources to excite the down-conversion films. And the down-conversion films consist of polymethyl methacrylate (PMMA) as polymer host and CdSe/ZnS QDs as color converters. We prepare flat and patterned QDs down-conversion films, and the patterned films are realized by transfer printing. The patterned down-conversion films with ordered and uniform microstructures can scatter the concentrated blue light emitted from TEOLEDs to suppress the viewing angle dependence. Thus, efficient and angle-stable white TEOLEDs are achieved.

2. Experimental

As shown in Fig. 1 (a), white TEOLED is fabricated by simply covering the blue TEOLED (BTEOLED) with down-conversion film. And BTEOLED is composed of Ag (100 nm)/MoO_3 (3 nm)/di-[4-(N,N-diptolyl-amino)-phenyl] cyclohexane (TAPC, 40 nm)/4,4′,4″-tris (carbazo-9-yl) triphenylamine (TCTA, 5 nm)/TCTA:bis(3,5-difluoro-2-(2-pyridyl)phenyl-(2-carboxypyridyl) iridium (III) (TCTA: 12 wt% Firpic, 25 nm)/1,3,5-tri[(3-pyridyl)-phen-3-yl] benzene (TmPyPB, 50 nm)/8-Hydroxy-quinolinolato-lithium (Liq, 2 nm)/Sm (20 nm)/TAPC (60 nm). The ITO-coated glass substrates are cleaned with Decon 90, and ultrasonically cleaned in deionized water baths for 15 min. After dried in the oven, the substrates are loaded in a vacuum evaporator for deposition. All devices are fabricated by thermal evaporating on the glass substrates under the high vacuum (6×10^{-4} Pa). The material deposition rate is monitored with quartz crystal. The typical deposition rate of organic materials and oxide materials is 0.05–0.15 nm/s, while that of metal materials is 0.15–0.25 nm/s. The current density-voltage-luminance characteristics and the spectra of the unpackaged devices are measured by using a Goniophotometric Measurement System (GP500, Otsuka Electronics Co. Osaka, Japan) in air at room temperature simultaneously.

The fabrication process of patterned down-conversion film is illustrated in Fig. 1 (b). The silicon wafer with concave microcylinder arrays on the surface is used as a substrate. The cylindrical hole is 5 μm deep and has a diameter of 20 μm. For the preparation of down-conversion film, a QD-PMMA mixture solution is obtained as follows: 0.285 g of PMMA is completely dissolved in 1 ml of toluene. And 100 μl of the above PMMA solution is blended with the QD toluene solution at a concentration of 100 mg/ml. The mixture solution is stirred for 10 min to avoid the separation of the QDs from the polymer matrix, because the aggregation of QDs would lead to luminescence quenching [30]. Then the viscous QD-PMMA mixture solution is poured cautiously onto the patterned Si substrate. And it is heated at 70 °C for 40 min to promote the solvent evaporation. The solidified down-conversion film is readily peeled off without damage, which is about 1 mm thick. And the patterns of the patterned Si substrate are transferred to the film. The fabrication process of flat down-conversion films is almost the same to that of the patterned ones, and the only difference is using flat Si substrate instead of the patterned substrate. Because thicker films bring more photons that trapped or guided within the films, which leads to

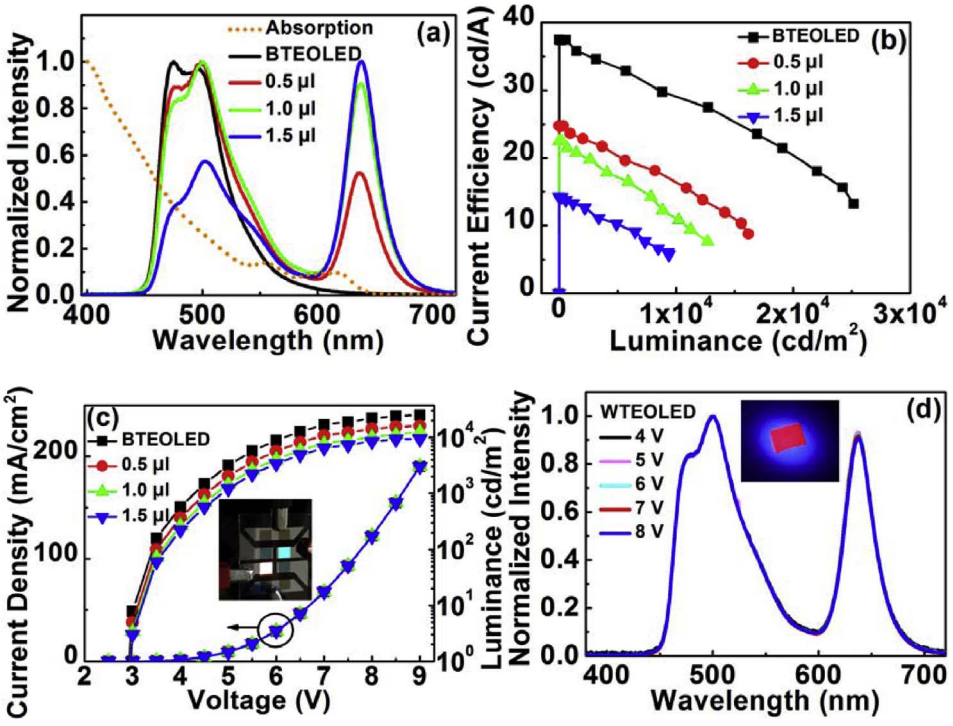

Fig. 2. (a) Normalized absorption spectrum of CdSe/ZnS QDs and EL spectra of BTEOLED and white TEOLEDs with the flat down-conversion films containing different volumes of QD solution (0.5, 1.0 and 1.5 μl). (b) Efficiency–luminance characteristics and (c) current density-voltage-luminance curves of BTEOLED and white TEOLEDs. The inset is the photograph of BTEOLED and WTEOLED. (d) Normalized EL spectra of WTEOLED under different driving voltages. The inset is the photograph of the flat down-conversion film containing 1.0 μl of QD solution under UV light.