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Highly efficient white transparent organic light emitting diodes with nano-structured substrate

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ABSTRACT

Enhancing outcoupling efficiency and stabilizing emission spectra are of high technical importance in realizing high quality white transparent organic light emitting diodes (TOLEDs). In this work, we demonstrate a random nano-scattering layer (RSL) as a structure which can effectively address those tasks. The RSL contributes to bottom and top emissions by scattering and reflection, respectively. With the use of RSL, we achieved remarkable total efficiency enhancement of 101%. Also, a viewing angle independent stable white spectrum with a color rendering index of 79 was achieved. With its straight forward processing, our RSL can be readily applied to deal with various photonic applications to enhance both efficiency and emission spectra.

1. Introduction

Organic light emitting diodes (OLEDs) provide high quality light that is environmentally friendly, and as a result OLEDs have emerged as preferred light sources for both flat panel displays and lighting applications [1–4]. By employing optically transparent components, transparent organic light emitting diodes (TOLEDs) have also been realized. When operating, TOLEDs emit light from both their bottom and top surfaces, while in non-operational mode they look like ordinary transparent glass. TOLEDs can be applied to a variety of window-like displays, bi-directional lighting facilities and eyewear displays [5–7].

In order to fully implement TOLEDs in real applications, several technical problems must be resolved. First, due to the asymmetric reflectivity of the transparent anode and semi-transparent cathode, the emission levels of the bottom and top of the TOLED are different. Usually the cathode emission is lower than that of the anode, which is typically comprised of Indium tin oxide (ITO). To solve this problem, a capping layer (CL) has been applied on the cathode surface to adjust the optical effect [8–11]. Second, the overall efficiency needs to be improved by employing an effective light outcoupling technique. Light outcoupling is crucial to achieve high luminance and long life time. Even though phosphorescent materials and carefully designed device structures have been used, optical losses still exist, which lessen the performance considerably. Typical loss modes include the total internal reflection at the substrate/air interface, and losses due to ITO/organic waveguide modes and surface plasmon polaritons (SPPs) at the metal electrode. Indeed, the light outcoupling efficiency of an OLED with flat glass does not exceed 20% [12–17].

Several methods have been suggested to improve light outcoupling. Examples include the micro cavity approach [18–20], micro lens array [21–24], and photonic-crystals structures [25–27]. In TOLEDs, the efficiency improving methods have involved using a low index capping layer (CL). However, many of these methods only result in limited enhancement and electroluminescence (EL) spectral distortion problems. To address these issues, scattering methods have been probed [28–32].

Here, we report the use of random scattering layers (RSL), previously developed by our group for single color OLEDs. We have fabricated white TOLEDs with highly enhanced light outcoupling using a RSL, which has the function of inducing scattering. The RSL was positioned between the ITO and glass substrate [33,34]. We

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investigated variations in the angular luminance distributions, efficiencies and electroluminescence (EL) spectra of the top and bottom emissions as a function of the random nano structure (RNS) thickness. We also studied the transmittance of the transparent metallic cathode and its effect on the performance of the white TOLEDs.

2. Experiments and simulations

The preparation of RSLs has been previously reported [33,34]. In order to obtain the RSLs, we used an array of randomly dispersed Ag droplets as a hard mask, and etched off the exposed region of a SiOx layer. The RNS consists of randomly distributed nano pillars with diameters of 50–700 nm (Fig. 1(b, c)). Nano pillar heights of 200, 300 and 350 nm were fabricated by changing the dry etching time. Then, a high refractive index-planarization layer (HRI-PL) of 300–800 nm thickness was formed on the RNS. The planarization layer had a high refractive index (n) of 2.0 at 550 nm. We call the composite layer, consisting of the RNS and a HRI PL, a RSL.

Fig. 1d shows the planarization layer. We used an organic/TiO_2-sol based hybrid material for the planarization film. A titanium oxide hydrate solution was prepared from a titanium(IV) butoxide in N,N-dimethyl acetamide (DMAc) solution. Polyvinylpyrrolidone (PVP, Mn¼ 40,000) was added to the Ti oxide-containing solution to adjust its viscosity. This mixture was spin-coated on the RNS. Subsequent baking at 400 °C yielded a very smooth (Ra < 1 nm) planarization film with a refractive index of 2.0. For details on the planarization materials, please refer to the following reference. [35,36] It is important to choose a planarization material which has higher n than ITO, otherwise a big fraction of the generated light will undergo total internal reflection [33,34].

All organic layers and the cathode were formed by a thermal evaporation method. Our white TOLEDs consisted of a stack structure of ITO (100 nm) 1,4,5,8,9,11-hexaazatriphenylene hexacarbonitrile (HAT-CN) (5 nm)/1,1-bis[(di-4-tolylamino)phenyl] cyclohexane (TAPC) (45 nm)/HAT-CN (10 nm)/TAPC (45 nm)/HAT-CN (10 nm)/TAPC (45 nm) (total HTL thickness:160 nm)/4,4′,4″-tris(N-carbazolyl)-triphenylamine (TCTA): Iridium(III)bis(4,6-difluorophenyl)-pyridinato-N,C2′)picolinate (Firpic)(7%) (5 nm)/ Red emission layer (0.5 nm)/Green emission layer (0.8 nm)/2,6-bis(3-(carbazol-9-yl)phenyl) pyridine (DCzPPy): Firpic(10%) (5 nm)/1,3-bis(3,5-di-pyrid-3-yl-phenyl)benzene (BmPyPB) (10 nm)/BmPyPB: Cs_2CO_3(45 nm: 20%)/LiF (1 nm)/Al (1.5 nm)/ Ag(20 nm)/CL(TAPC: 110 nm). We fabricated the TOLEDs with an emitting area of 10 mm × 7 mm. The base pressures of all deposition processes were below 6.66×10^{-5} Pa. Prior to the evaporation process the ITO surface was treated by oxygen plasma to remove contaminants. The fabricated TOLED devices were glass encapsulated immediately in an inert glove box environment. UV curable epoxy resin was used to glue the glass, and a getter was included to collect moisture.

Transmittance of the glass encapsulated TOLED was measured using an UV–visible spectrophotometer (U-3501, Hitachi). The electroluminescence (EL) spectra and the angular luminance distributions were collected using a goniometer equipped spectroradiometer (CS-2000, Minolta). The current density (J) –voltage (V) –luminance (L) characteristics were obtained using source\measure units (Keithley 238) and the aforementioned spectroradiometer. The surface morphologies of the cathode films were investigated by means of scanning electron microscopy (SEM, Model: Sirion 400, Philips). The main input variables were the refractive index (n), extinction coefficient (k) and film thickness. In order to obtain realistic simulation results, we used all measured values of n and k of the organic materials, as determined using an ellipsometer (M-2000D, J.A. Woollam Co.). Additionally, transmission haze was measured using a haze meter (Haze-gard plus, BYK Additives & Instruments).

3. Results and discussions

Fig. 1(a) shows the cross-sectional structure of our white TOLED. A three-color type OLED was adopted to obtain white light. Indium tin oxide (ITO) and LiF/Al/Ag were used as the transparent anode and transparent cathode, respectively. In order to enhance the transmittance or the efficiency of our white TOLED, we equipped it with a CL of 1,1-bis[(di-4-tolylamino)phenyl]cyclohexane (TAPC) on the top of the cathode. The optical effects of the CL on TOLED performances have been investigated in detail [9–11]. For the hole transport layer (HTL), we used an alternating structure of Hat-CN and TAPC [37,38]. Such an HTL structure has been demonstrated

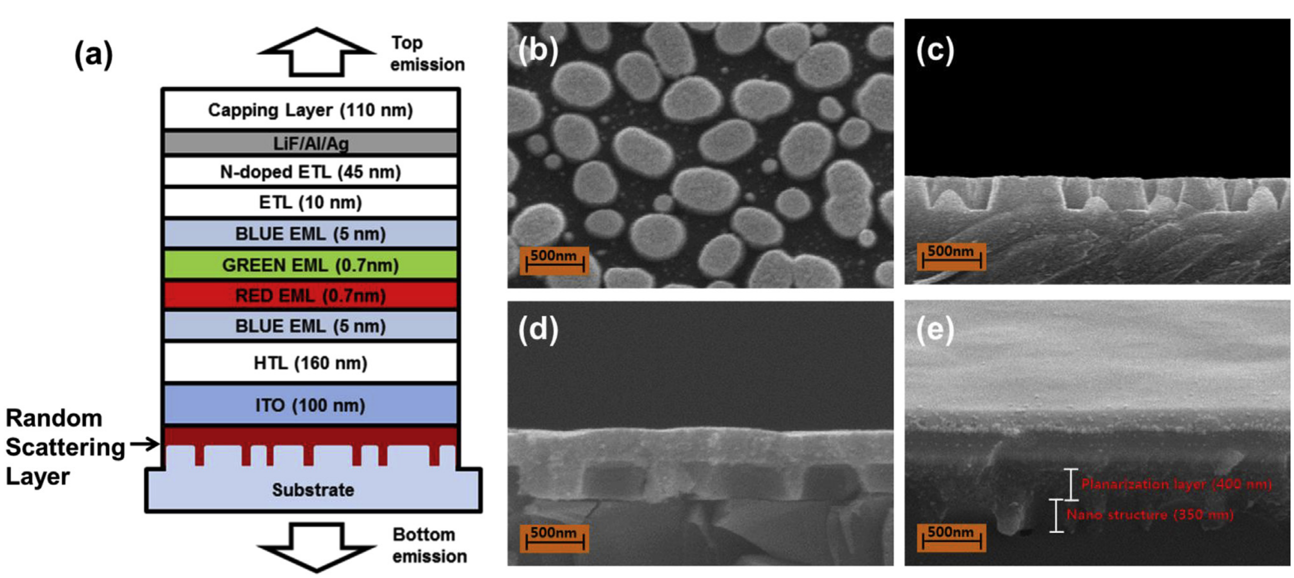

Fig. 1. (a) Schematic of the white TOLED equipped with RSL, SEM image of (b) irregular nano-sized pillars(top view), (c) irregular nano-structures(cross section), (d) random scattering layer with planarization layer (cross section), (e) TOLEDs(cross section 45°).