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High-efficiency and superior color-stability white phosphorescent organic light-emitting diodes based on double mixed-host emission layers

Ping Chen*, Bingye Chen, Liangmei Zuo, Yu Duan, Guangguang Han, Ren Sheng, Kaiwen Xue, Yi Zhao

State Key Laboratory on Integrated Optoelectronics, College of Electronic Science and Engineering, Jilin University, Changchun 130012, People's Republic of China

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ABSTRACT

We demonstrate high-efficiency and superior color-stability white phosphorescent organic light-emitting diodes based on double blue mixed-host emission layers (EMLs) with different mixed ratios. The key feature of the concept is to introduce double blue mixed-host EMLs with an orange ultrathin layer sandwiched between them. The improved white device without spacer or interlayer achieves superior color-stability and reduced efficiency roll-off, which are consistent with the good ambipolar conductivity of the mixed-host layer. Moreover, peak efficiency of 40.8 lm/W and low turn-on voltage of 2.71 V are realized. The double mixed-host EMLs concept proves to be quite useful in achieving excellent device performance.

1. Introduction

During the past two decades, white organic light-emitting diodes (OLEDs) have been intensively developed due to their potential applications in backlight, full color displays and lighting sources for general illumination [1–4]. High efficiency, low cost and good color stability of white OLEDs are required to meet widespread commercial use [5,6]. In order to achieve the goal, phosphorescent materials those could harvest both singlet and triplet excitons are often used as emitters, achieving an internal quantum efficiency of 100% in principle [7]. To generate white light emission, three primary or two complementary colors emitters are usually incorporated and distributed in single or multiple emission layers (EMLs) [8–10]. As we all know, the distribution of carriers and excitons decides the performance of white OLEDs, thus the doping concentration and the thickness of EMLs should be carefully adjusted to obtain balanced white light. For single EML white OLEDs, the doping concentration of orange or red emitters must be accurately controlled at a very low level (~0.5%), which increases manufacturing complexity and raises the commercialization cost. In contrast, it's relatively easier for multiple EMLs white OLEDs to control the doping concentration and achieve high efficiency. However, it is very difficult for multiple EMLs white OLEDs to obtain stable white light emission, which especially hampers the application of white OLEDs in lighting.

There are many influencing factors for multiple EMLs white OLEDs on color stability: carrier trapping [11], shift of exciton recombination zone [12,13] and triplet–polaron annihilation or triplet–triplet annihilation [14,15]. To alleviate the issue, some researchers begin to insert spacer or interlayer with ambipolar charge carrier transport properties between EMLs. For example, to fabricate color-stable hybrid white OLEDs using exciton blocking interlayer, Schwartz et al. investigated the charge carrier mobility of mixed interlayer of N,N-di(naphthalen-1-yl)-N,N-diphenyl-benzidine and bis(2-methyl-8-quinolinato) -4-phenylphenolate aluminum with different mixed ratios [16]. D'Andrade et al. introduced a multi-emission layer fully electrophosphorescent white OLEDs with exciton blocking layers 4,4-N,N-dicarbazole-biphenyl between EMLs, which achieved Commission International de L'Eclairage (CIE) coordinates of (0.35, 0.36) and a maximum current efficiency of 6.1 ± 0.6 cd/A [17]. Zhao et al. reported efficient white

* Corresponding author.
E-mail address: pingchen@jlu.edu.cn (P. Chen).

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device with 4,4′,4″-tri(N-car-bazolyl)-triphenylamine and bis[2-(2-hydroxyphenyl)-pyridine] beryllium as mixed interlayer. The hybrid white device with excellent stable spectra exhibited high efficiencies of 30.2 cd/A and 32.0 lm/W [18]. However, this kind of device structures with spacer or interlayer complicates the fabrication process and increases the fabrication cost, which is not helpful for mass production. Additionally, the driving voltage always increases and the power efficiency generally decreases due to the presence of heterojunction interfaces by the additional layer. Thus, it is necessary to develop color-stability white OLEDs without spacer or interlayer.

In this paper, we present a novel design of multiple EMLs device structure without spacer or interlayer to achieve stable and efficient white light emission. Our designed white device contains double blue mixed-host EMLs and an ultrathin non-doped orange EML. The key feature of our concept is that both the two blue EMLs adopted mixed-host system composed of hole-transport material (HTM) and electron-transport material (ETM) with different mixed ratios. The mixed ratio of HTM to ETM in the mixed-host EMLs are assigned 2: 1 near the hole-transport layer (HTL) and 1: 2 near the electron-transport layer (ETL), respectively. So the interfacial energy barriers at the EMLs interfaces are further reduced, which effectively facilitates the carrier transport and expands the exciton recombination zone. One optimized white device exhibits high efficiencies of 40.8 lm/W and 39.8 cd/A with negligible CIE coordinates change of (0.005, 0.008) during a wide range of luminance. In addition, the white device also shows low turn-on voltage and reduced efficiency roll-off.

2. Experimental

The chemical structures and detailed energy level diagram of the materials proposed are shown in Fig. 1. All the devices were fabricated on indium-tin-oxide (ITO)-coated glass with a sheet resistance of 20 Ω/sq. 2 nm Molybdenum(VI) Oxide (MoO_3) was used as hole-injection layer. 4,4′,4″-tri(N-car-bazolyl)-triphenylamine (TCTA) was used as HTL, and 2,2′,2″-(1,3,5-Benzinetriyl)-tris(1-phenyl-1-H-benzimidazole) (TPBi) was used as ETL. Here, TCTA with high hole-transport mobility and TPBi with high electron-transport mobility were blended as the mixed-host for blue phosphorescent emitter. The phosphorescent emitters for the orange and blue emission were Iridium(III) bis(4-phenylthieno[3,2-c]pyridinato-N,C2′) acetylacetonate (PO-01) and iridium (III) Bis(3,5-diflouro)-2-(2-pyridyl)phenyl-(2-carboxypyridyl) (FIrPic). 100 nm Al modified by 1 nm Liq was used as cathode. Thermal evaporation of organic materials was carried out in a high vacuum condition without breaking vacuum (~10^{-7} torr). All the layer thicknesses and the deposition rate of materials were calibrated with an oscillating quartz thickness monitor. Electroluminescence (EL) spectra and CIE coordinates of the devices were measured by using a PR655 spectroscan spectrometer. Current-Voltage-Luminance characteristics of the fabricated devices were measured by combining the spectrometer with a programmable Keithley 2400 voltage–current source in air at room temperature.

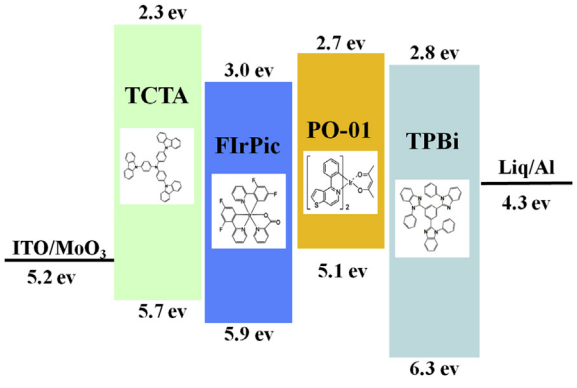

Fig. 1. The detailed energy level diagram and chemical structures of the materials.

3. Results and discussion

Compared to the green and red counterparts, blue phosphorescent OLEDs have lower efficiency and shorter lifetimes and need to be further improved [19]. In order to prevent the rapid exothermic transfer of energy, the host material for blue emitters should possess higher triplet excited state. However, a high triplet excited state will cause a series of problems such as high carrier injection barrier at the EMLs interfaces and high driving voltage. And large densities of charge carriers pile up at the EMLs interfaces, resulting in low efficiency and severe efficiency roll-off. Here, we introduced double mixed-host EMLs in blue OLEDs to further improve the performance. The designed device structure of double mixed-host EMLs facilitates the carrier injection and transport and expands the exciton recombination zone, which leads to charge balance and lower driving voltage. In order to clearly understand the effect of adopting double mixed-host EMLs in blue OLEDs, structural comparison in EML has been carried out in the following part, as depicted in Fig. 2(a). Devices A and B utilize TCTA or TPBi single host doped with 10% FIrPic. Device C utilizes single mixed-host EML, while device D consists of double mixed-host EMLs with the mixed ratio of TCTA to TPBi are assigned 2: 1 near the HTL and 1: 2 near the ETL, respectively. The mixed-host EMLs consist of TCTA and TPBi uniformly doped with 10% FIrPic. Fig. 2(b) shows the current density-voltage characteristics of the devices A-D. The lower current density for devices A and B may be the result from the high energy barrier between EML and transport layers that obstructs the charge transport. Device D using double blue mixed-host EMLs with bipolar transport property shows the highest current density, which indicates better charge injection and transport in the EMLs.

The normalized EL spectra of the blue devices A-D at 4 V are shown in Fig. 2(c). All the devices show a primary emission peak at 476 nm and a subpeak at 496 nm originating from FIrPic. The performances of blue OLEDs A-D are summarized in Table 1. It is found that the peak power efficiency of 21.56 lm/W is realized in device D. In devices A and B, TCTA with electron block characteristics and TPBi with hole block characteristics are used as host materials respectively, having poor overall transport properties for either electrons or holes. Hence, it is difficult to achieve balanced carrier injection and transport in EML, leading to lower efficiency. While devices C and D based on mixed-host structure could achieve more balanced carrier injection and broader recombination zone due to the bipolar characteristics of EMLs. Furthermore, seen from Fig. 2(d), devices A and B with single host EML show much higher turn-on voltages (at measureable luminescence of 1 cd/m^2) of 3.45 V and 3.82 V, respectively. For devices A-B, the higher turn-on voltages are largely due to the higher interfacial energy barriers at the interface of EML/TPBi or TCTA/EML for electrons or holes injection. Compared to other devices, device D shows the lowest turn-on voltage of 2.83 V, which is understandable since the energy barriers at the EMLs interfaces are further reduced. Double EMLs [20] and mixed host systems [21] have been employed to improve device lifetime. We believe the designed device structure with double mixed-host EMLs is beneficial for the improvement in lifetime due to the reduced interfaces barriers and balanced charge carriers. Further research and analysis will be carried out in our future research work.