



Effect of exciton blocking layers on the color-tunable properties of organic light-emitting devices



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ABSTRACT

A series of voltage-controlled color-tunable organic light-emitting devices (OLEDs) were fabricated. Four charge transport materials with different characteristics serving as the exciton blocking layers (EBLs) were inserted between yellow and blue emissive layers, respectively, and the color-tunable properties of OLEDs were studied in detail. Carrier transport and electroluminescence (EL) mechanisms of the color-tunable devices were investigated by analyzing the EL spectra and device efficiencies. The results showed that a color-tunable range of blue, white, and yellow light had been realized by tuning the film thicknesses of EBLs. The device with 16 nm *N,N'*-dicarbazolyl-3,5-benzene hole transport material as the EBL could shift the color coordinates from (0.21, 0.32) to (0.38, 0.41), which exhibited the broadest color tuning range. Meanwhile, the device based on 18 nm 1,3,5-tris(*N*-phenylbenzimidazol-2-yl)benzene electron transport material had the opposite color-tuning sequence and slightly higher tuning voltages, comparing with other three devices. The difference of color-tunable properties in OLEDs based on various EBLs was attributed to the variation of carrier mobility and energy level of EBL materials. Moreover, to effectively harvest the excitons for color-tunable light emission, the EBL thickness should be comparable to the triplet exciton diffusion length of EBLs.

1. Introduction

Organic light-emitting devices (OLEDs) have drawn considerable interests for their broad applications in full-color flat-panel displays and solid state light sources [1–5]. Along with the trend of OLEDs evolving into a mature technology in recent years, many newly-designed devices have emerged for practical environmental friendly and human healthy applications [6], such as candle light-style OLEDs and sunlight-style chromaticity tunable OLED [7,8], color of light shows a marked effect on human health and the growth of crops [9]. For instance, cold white light is helpful to keep people awake and active [10]. While yellow light is conducive to the growth of plant height [11], and blue light is beneficial to the protein synthesis [12]. Therefore, the voltage-controlled color-tunable OLEDs, which can generate two or more colors with a simple adjustment of bias voltage in one device, are of particular attention in the fields of decoration or agriculture industry due to their inherent advantages.

The previous works to realize color-tunable OLEDs mainly focused on the innovation of device structure and the realization of novel functional materials. For example, the first demonstrated integrated color-tunable OLEDs made use of stacked pixel architecture, allowing

for independent tuning of color and intensity of each pixel [13]. Mg-Ag-ITO ultra-thin electrode was used in this device as the hole injection contact for a red color emitting device stacked on top of one transparent blue emitting OLED. Analogously, in the aspects of structure optimization, inserting Al/WO₃/Au alloy metal or Ag/Au thin film as transparent common electrode in the stacked emitting structure [14,15], and using side-by-side structure devices fabricated by alignment free mask patterning method [16], were also approaches to realize voltage-controlled color-tunable OLEDs. In addition, some novel materials, including lanthanide metal-organic framework materials [17], β-diketone complexes [18], gold(III) complexes [19], and DNA/PAN complex et al. [20], were reported to achieve this goal.

However, the above-mentioned methods are involved with the complicated device structure and timing consuming novel material design and synthesis. Alternatively, as low cost and simple structure are becoming more and more important in the consideration of designers, inserting an exciton blocking layer (EBL) between two or more emitting layers (EMLs) to realize different color emissions in single device structure, which reduces the fabrication complexity and cost, is an easier and more effective way to fabricate color-tunable OLEDs [21]. For example, Jou et al. realized a sunlight-style color-temperature

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tunable OLED [8], by using an interlayer to regulate the distribution of holes. Our group also introduced a simple architecture of color-tunable OLEDs by inserting an exciton adjusting layer between dual EMLs [22,23]. Nevertheless, these previous works mainly aimed at how to initially realize the color-tunable OLEDs with only one kind of interlayer, and a further systematic comparative work of the impact caused by different interlayers on color-tunable properties had not been studied. Therefore, there is still lack of research on the deep insight in the influence of different interlayer materials with various carrier mobilities and energy levels on the color-tunable properties including color-tuning range, sequence and tuning voltage of OLEDs.

In this work, a series of color-tunable OLEDs with emission tunability including blue, yellow and white region were fabricated, by using four different EBLs as the interlayer inserted between yellow and blue EMLs. For comparison, three hole transport materials including *N,N'*-dicarbazolyl-3,5-benzene (mCP), *N,N'*-bis(3-methylphenyl)-*N,N'*-diphenylbenzidine (TPD), 1,1-bis[(di-4-tolylamino)phenyl]cyclohexane (TAPC) and one electron transport material of 1,3,5-tris(*N*-phenylbenzimidazol-2-yl)benzene (TPBi) were selected as the EBLs, respectively, to systematically contrast the performance differences between P-type and N-type EBLs based devices. By optimizing the film thickness of EBL, color-tunable OLEDs with different color-tuning ranges, sequences and tuning voltages based on these four EBL materials were realized. To explain the difference of color-tunable principle of devices with different EBLs and film thicknesses, the transporting mechanism of carriers and excitons in color-tunable devices had been studied from the perspectives of carrier mobility and energy level of EBL materials. Also, the relationship between the optimized EBL thickness to achieve color-tunable OLEDs and the triplet exciton diffusion length of EBL material was discussed.

2. Experimental

Indium tin oxide (ITO) coated glass substrates with a sheet resistance of 15 Ω /sq. were cleaned in detergent water, acetone, deionized water and isopropyl alcohol by using an ultrasonic bath for 15 min, respectively. Thereafter, the pre-cleaned ITO substrates were dried with nitrogen gas flow and then, they were treated with O_2 plasma in a vacuum chamber with a pressure of 25 Pa for 5 min, to clean the surface and increase work function. Organic functional and metallic layers were deposited in different chambers under high vacuum conditions of less than 3×10^{-4} and 3×10^{-3} Pa, respectively. Deposition rates and thicknesses of thin films were monitored by using a quartz crystal oscillator. Device area was controlled by shadow mask, and the typical value was 0.2 cm². Luminance-current density-bias voltage (*L*-*J*-*V*) characteristics curves were tested by a Keithley-4200 semiconductor characterization system and luminance meter ST-86LA. Electroluminescence (EL) spectra and the Commission International de l'Éclairage (CIE) coordinates of devices were measured and analyzed by an OPT-2000 spectrophotometer. All measurements were performed in air at room temperature without device encapsulation.

As shown in Fig. 1, the configuration of color-tunable OLEDs in this work is as follows: ITO/NPB (45-X nm)/(t-bt)₂Ir(acac) (1 nm)/EBLs (X nm)/UGH2: FIr6 (10 wt.%, 16 nm)/Bphen (32 nm)/Mg:Ag (100 nm). The devices were composed of two EMLs, a blue EML with phosphorescent iridium(III) bis(4',6'-difluorophenylpyridinato)tetrakis (1-pyrazolyl)borate (FIr6) doped in *p*-bis(triphenylsilyl)benzene (UGH2) with high luminous performance [24], and an ultrathin non-doped yellow EML using phosphorescent bis[2-(4-*tert*-butylphenyl)benzothiazolato-*N,C*^{2'}]iridium(acetylacetonate) [(t-bt)₂Ir(acac)] with remarkable hole-trapping ability and narrow band-gap [25], which are benefit for exciton transfer from blue emitter to obtain obvious yellow emission. *N,N'*-diphenyl-*N,N'*-Bis(1-naphthyl-phenyl)-(1,1'-biphenyl)-4,4'-diamine (NPB) and 4,7-diphenyl-1,1'-phenanthroline (Bphen) were used as hole transport layer (HTL) and electron transport layer (ETL), respectively. An electron transport material TPBi, three hole transport materials

TAPC, TPD and mCP were chosen as EBLs, which would be inserted between the yellow and blue EMLs. The thicknesses of EBLs would be adjusted to analyze their effect on device performance.

3. Results and discussion

Fig. 2 and 3 show the relationship between the EL spectra, EBL film thickness and bias voltage of all OLEDs. It is obvious that each EL spectrum can be divided into two parts. The blue emission component with a 460 nm peak and a 490 nm shoulder is ascribed to FIr6 [26], and the yellow emission component with a 563 nm peak and a 600 nm shoulder is originated from (t-bt)₂Ir(acac). It can also be seen that there is a variation in the ratio of blue to yellow emission intensity in different devices. The EL spectra of devices with thin EBLs are all dominated by yellow emission from (t-bt)₂Ir(acac), while the blue emission intensity of FIr6 increases with the enhancement of EBL thickness.

This phenomenon can be explained as follows: there are three exciton formation regions located at two EMLs and one EBL. Excitons formed in EBL are anticipated to diffuse and be injected into the neighboring EMLs, and recombine to yield blue and yellow emissions. The devices with thin EBLs have a same exciton transfer mechanism, in which a majority of excitons are transferred from FIr6 to (t-bt)₂Ir(acac), due to the narrower energy band-gap of (t-bt)₂Ir(acac). With the increase of EBL film thickness, the excitons transfer from blue to yellow EML is suppressed, and inhibited finally, owing to the enhanced effect of EBLs on exciton blocking. As a result, an approximate independent emission intensity of the blue and yellow components was obtained, and the devices with thick EBL exhibit high color stability by changing bias voltage. In other word, the OLEDs don't have color-tunable property.

When the EBL film thickness is optimized, the color-tunable devices can be achieved, as an optimum amount of excitons is allowed to transfer between two EMLs. From the EL spectra of Fig. 2(c), (f) and (j), it is clear that the devices composed of 16 nm-thick mCP, 8 nm-thick TPD and 4 nm-thick TAPC EBLs show an excellent color-tunable capability. These three devices possess the most voltage-sensitive EL spectra consisting of white, blue, and yellow light. This phenomenon is attributed to the sensitive exciton transfer zone generated by EBLs with proper film thickness, responsible for a wide range of emission color at different biases.

In the devices using hole transport materials of mCP, TPD and TAPC as EBLs, a wide range color emissions with the tuning sequences from blue, white, to yellow light is obtained, by varying the bias voltage from 6 to 10 V, which is relevant to the favorable hole transport features of these materials. The energy barriers of 1.0, 0.7 and 0.6 eV for mCP, TAPC and TPD, respectively, for hole injection into EBL, are lower than those of electron injection of 1.4, 1.7 and 1.3 eV for the three materials. More charge recombination in blue EML will occur accordingly. At a low bias voltage of 6 V, the blue emission intensity in these devices is much stronger than their yellow counterpart, and the OLEDs exhibit blue color. Then with the increase of bias voltage, more charges recombine in (t-bt)₂Ir(acac) layer, and the yellow emission intensity is enhanced to achieve white light. With the further increase of yellow emission intensity, OLEDs with yellow light emission are observed at high bias voltage.

As shown in Fig. 3(c), for the device with an electron transport material of TPBi as EBL, a similar color-tunable range to that of hole transport materials is obtained. In contrast, the tunable sequence with the increase of voltages is opposite to the above three materials. In this device, an electron injection barrier of 1.0 eV is much lower than that of hole injection of 1.4 eV, and the mobility of electron is much higher than that of hole in TPBi. Hence, more holes and electrons combine with each other in (t-bt)₂Ir(acac) layer. With the increases of bias voltage, the trapping effect of (t-bt)₂Ir(acac) will be suppressed [27]. Thus as the voltage further increased, more exciton will remain in blue EML, the blue portion becomes predominant because of the high efficiency of

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