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Energy Procedia 12 (2011) 620 – 624

Energy

Procedia

ICSGCE 2011: 27–30 September 2011, Chengdu, China

Influence of Doping Concentrations in White Organic Light-emitting Diodes based on Phosphorescent Iridium Complexes

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Abstract

Phosphorescent white organic light-emitting diodes (PhWOLEDs) based on blue (Firpic) and yellow [(t-bt)2Ir(acac)] phosphorescent emitters with various doping concentration were reported. A PhWOLED with high performance characteristics has been obtained when the doping concentration for Firpic and (t-bt) 2Ir (acac) is 8 wt% and at 6 wt%, respectively. The Commission Internationale del'Eclairage (CIE) coordinates of the optimal PhWOLED is (0.333, 0.416) at 10 V. The high performance of the optimized PhWOLED is attributed to effective host-guest energy transfer and efficient exciton recombination combining with suppressed exciton-quenching process in the emitting layers.

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Selection and/or peer-review under responsibility of University of Electronic Science and Technology of China (UESTC)

Keywords: Phosphorescent white organic light-emitting diodes; Doping concentration; Charge-trapping

1. Introduction

White organic light-emitting diodes (WOLEDs) have attracted much attention due to their potential applications in next generation displays and solid state lighting [1]. Recently, the efficiency of WOLED has been dramatically improved, which is beneficial for saving energy consumption [2]. This progress will certainly speed up the utilization of OLEDs in lighting in the near future. Therefore, the development of OLEDs can play an important role in solving the global energy crisis.

To achieve WOLEDs with high efficiency and brightness, electrophosphorescence stands out as one of the most effective mechanisms since they can utilize both singlet and triplet excitons for generating light emission, leading to OLEDs with 100 % internal quantum efficiency (IQE) [3]. Phosphorescent WOLEDs have recently attained a power efficiency up to 40 lm/W based on the strategies of three phosphorescent

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red-green-blue (RGB) emitters in combination with a p-i-n device structure (i.e. using both p- and n-types doped transporting layers) [2] Another common approach for making WOLEDs involves the simple use of the combination of two complementary colors such as blue and yellow. The main advantages of such devices are their much simpler structures and more stable colors. As for device fabricating, dye doping is a promising way to improve the performance of organic light-emitting diodes (OLEDs) [4]. The doping method prevents both concentration quenching of the dyes' emission, and crystallization of the dye molecules [5]. Therefore, OLEDs fabricated by the dye doping method generally exhibit higher efficiency, excellent stability, and even good spectral purity.

Here, we report white OLEDs with high efficiency and brightness based on the phosphorescent emitters iridium (III) bis (4, 6-(di-fluorophenyl)-pyridinato-N, C2') picolinate (Firpic) and [2-(4-tertbutylphenyl) benzothiazolato-N, C2'] iridium (acetylacetonate) [(t-bt)₂Ir(acac)]. N, N'-dicarbazolyl-3, 5-benzene (mCP) doped with Firpic and p-bis(triphenylsilyl)benzene (BPhen) doped with (t-bt)₂Ir(acac) were used as blue and yellow emitting layers, respectively. Firpic has been widely used as an efficient blue phosphor [6]. The combination of these two emitters results in a PhWOLED with high efficiency and brightness. The influence of doping concentration of the dopants on the device performance was discussed.

2. Experimental Details

Figure 1 shows the molecular structures of the organic materials used in this study. The novel phosphorescent material (t-bt)₂Ir(acac) was synthesized in our laboratory[7-9]. The organic material of TAPC was purchased from Sigma-Aldrich Co. The organic materials of mCP, Firpic and BPhen were purchased from LumTec Co without further purification.

Indium-tin-oxide (ITO)-coated glass with a sheet resistance of 10 Ω/sq was employed as anode substrate. Before thermal deposition, ITO glasses were ultrasonically cleaned with detergent water, acetone, de-ionized water, and ethanol for 15 minutes at each step, and then dried in nitrogen gas flow. Afterwards, cleaned ITO substrates were treated by oxygen plasma for 5 min to enhance the work function of ITO, and then loaded into an organic vacuum evaporation chamber. The organic films were deposited at a rate of 0.1~0.5 Å/s under a pressure of 3×10⁻⁴ Pa, the metallic cathode was deposited at a rate of 1~5 Å/s under a pressure of 3×10⁻³ Pa without breaking the vacuum. The deposition process was *in situ* monitored using a quartz crystal oscillator mounted close to substrate holder. Current density-voltage-luminance (*J-V-L*), electroluminescent (EL) spectral characteristics of the devices were measured with KEITHLEY-4200 semiconductor characterization system and an OPT-2000 spectrophotometer. All the measurements were performed at room temperature in ambient circumstances. The typical emission area of all devices was 5 mm × 4 mm.

The optimized device structure was ITO/ 1, 1-bis-(4-bis (4-tolyl)-aminophenyl) cyclohexene (30 nm)/ mCP: 8 wt% Firpic (20 nm)/ BPhen: 6 wt% (t-bt)₂Ir(acac) (15 nm)/ BPhen (40 nm)/ Mg:Ag (200 nm, the ratio of Mg to Ag is 10: 1). Here TAPC was used as a hole-transporting layer (HTL) and exciton-blocking layer (EBL). BPhen was used as an electron- transporting layer (ETL). Mg:Ag was used as the cathode.

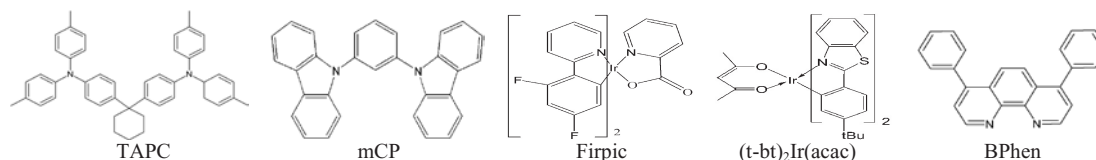


Fig.1. Molecular structures of organic materials in this work

3. Results and Discussion

Since we focus on PhWOLEDs, prior investigations on the OLED structures incorporating the blue phosphorescent emitters are necessary. We first fabricated blue OLEDs using mCP doped Firpic as the EML. The concentration of Firpic in mCP is varied.

Type B: ITO/TAPC (30 nm)/mCP: x wt% Firpic (20 nm)/BPhen (40 nm)/Mg:Ag(200 nm) (x = 4, device B1; x = 8, device B2; x = 16, device B3).

Figure 2 shows the EL spectra of these devices. It can be seen that the major peaks of these devices locate at 470 nm and the shoulder peaks locate at 493 nm, ascribed to the emissions from metal to ligand charge transfer and π - π^* transitions, respectively, in the Firpic guest molecules [10]. It indicates the effective energy transfer from mCP host to Firpic guest [11].

Figure 3 shows the current efficiencies and power efficiencies of our blue Firpic-doped OLEDs (devices B1-B3). The maximum luminance, luminous efficiency and power efficiency of device B2 are 15,894 cd/m^2 , 9.2 cd/A and 5.1 lm/W , respectively, which are the highest values among the three devices. Insufficient molecules are involved in the process of light emission when Firpic concentration is 4 wt%, resulting in lower luminance and efficiencies. The device performance is improved at 8 wt% Firpic concentration as a result of enhanced recombination of triplet excitons on Firpic dopant. However, the device performance slightly degraded at 16 wt% Firpic concentration due to the concentration quenching associated with the increased molecules. So the optimal doping concentration for Firpic is at 8 wt% in this device structure.

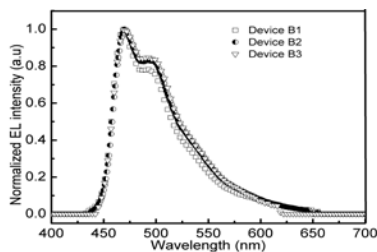


Fig. 2. Normalized EL spectra of devices B1-B3 at the applied voltage of 10 V.

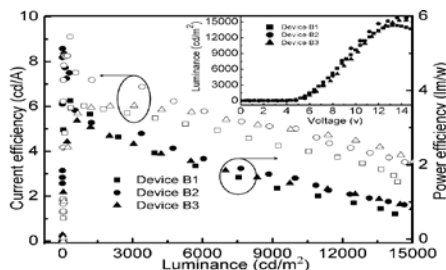


Fig. 3. Current and power efficiencies of devices B1-B3. Inset: the voltage-luminance characteristics of device B1-B3.

Therefore, the Firpic doping concentration was fixed at 8 wt% to investigate the influence of doping concentration of $(t\text{-}bt)_2\text{Ir}(\text{acac})$ on the performance of the two-color PhWOLEDs. The configurations include the following devices.

Type W: ITO/TAPC (30 nm)/mCP: 8 wt% Firpic (20 nm)/BPhen: x wt% $(t\text{-}bt)_2\text{Ir}(\text{acac})$ (15 nm)/BPhen (40 nm)/Mg:Ag(200 nm) (x = 2, device W1; x = 6, device W2; x = 10, device W3).

Figure 4 shows the current density-voltage-luminance characteristics of the PhWOLEDs (device W1-W3) with different doping concentration of $(t\text{-}bt)_2\text{Ir}(\text{acac})$. It can be seen that the current density decreases with increasing concentration of $(t\text{-}bt)_2\text{Ir}(\text{acac})$ dopant, which indicates that $(t\text{-}bt)_2\text{Ir}(\text{acac})$ molecules should play a role of hole charge trapping due to the higher-lying HOMO (highest occupied molecular orbital) (5.2 eV) of $(t\text{-}bt)_2\text{Ir}(\text{acac})$ with respect to the HOMO (6.5 eV) of Bphen [12]. In the case of direct charge trapping, the dopant molecules may be considered as the shallow trapping centers, which will trap injected charge carriers and change the charge density, resulting in a dependence of current-density-voltage (J-V) characteristics on the doping concentration [13]. Device W2 exhibits higher luminance than that of the other two devices due to efficient exciton recombination in the emitting layers.

Figure 5 shows the current efficiencies and power efficiencies of devices W1-W3. In the case of

device W1, few holes are trapped then few excitons would be formed on the (t-bt)₂Ir(acac) guest molecules for its lower doping concentration. The device performance is enhanced when the doping concentration of (t-bt)₂Ir(acac) increased to 6 wt%. A large amount of excitons on (t-bt)₂Ir(acac) are generated as the doping concentration of (t-bt)₂Ir(acac) increased to 10 wt%, which is likely to trigger the occurrence of exciton-quenching [14] [15], resulting in poor device performance compared with device W2. It demonstrates that 6 wt% is the saturated concentration of (t-bt)₂Ir(acac) in this series of devices. It is interesting that the PhWOLEDs exhibit much higher luminance and efficiencies than that of the single Firpic-doped blue OLEDs, which is attributed to effective host-guest energy transfer and efficient exciton recombination on the (t-bt)₂Ir(acac) dopant combining with suppressed exciton quenching.

Warm white emission has been obtained in the three devices. Fig. 6 shows the normalized EL spectra of the optimized white light-emitting device at the voltage of 5 V. The spectra contains two main peaks located at 471 and 561 nm, which are ascribed to light emission from Firpic in the blue emissive layer and that from (t-bt)₂Ir(acac) in the yellow emissive layer. The Commission Internationale del'Eclairage (CIE) of device W2 is (0.333, 0.416) at 10 V.

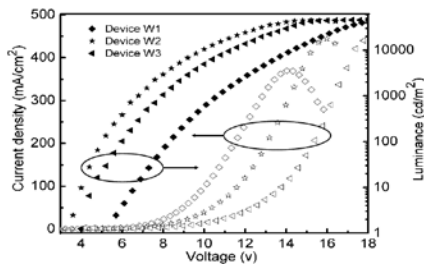


Fig. 4. The current density-voltage-luminance characteristics of devices W1-W3.

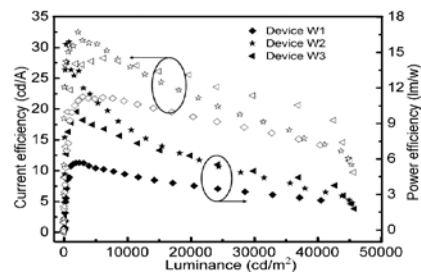


Fig. 5. Current and power efficiencies of devices W1-W3.

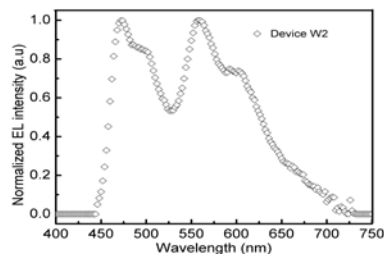


Fig. 6. Normalized EL spectra of device W2 at 5 V.

4. Conclusion

We have demonstrated high luminance and efficiencies PhWOLED with a structure of ITO/TAPC (30 nm)/mCP: 8 wt% Firpic (20 nm)/BPhen: 6 wt% (t-bt)₂Ir(acac) (15 nm)/BPhen (40 nm)/Mg:Ag.

We have studied the EL characteristics of single Firpic-doped blue OLEDs and two-color PhWOLEDs by varying the doping concentration of the phosphorescent emitters.

Acknowledgements

This work was supported by the National Science Foundation of China (NSFC) via Nos. 60736005 and 60425101-1, the Foundation for Innovation Groups of NSFC via No. 61021061, Fundamental Research Funds for the Central Universities (Grant No. ZYGX2010Z004), SRF for ROCS, SEM via No. GGRYJJ08-05 and Doctoral Fund of Ministry of China via No.20090185110020.

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