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Highly efficient green and red phosphorescent OLEDs using novel bipolar blue fluorescent materials as hosts

J. Xiao^{a,*}, Z.Y. Lü^b, X.K. Liu^c, Y.F. Han^d, L.R. Zheng^a

^a College of Physics and Electronic Engineering, Taishan University, Tai'an, Shandong 271021, PR China

^b Department of Physics, School of Science, East China University of Science & Technology, Shanghai 200237, PR China

e Nano-Organic Photoelectronic Laboratory and Key Laboratory of Photochemical Conversion & Optoelectronic Materials, Technical Institute of Physics &

Chemistry, Chinese Academy of Sciences, Beijing 100190, PR China

^d Department of Chemistry and Chemical Engineering, Taishan University, Tai'an, Shandong 271021, PR China

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ABSTRACT

We investigated highly efficient green and red phosphorescent organic light-emitting diodes (OLEDs) based on two kinds of iridium complexes as the guests and two novel 1,3,5-triazine derivatives as the host materials, respectively. For comparison, the devices using a common fluorescent host 4,4'-*N*,*N*'-dicarbazolebiphenyl (CBP) have also been fabricated. Results show that all devices using 1,3,5-triazine derivatives as host have better performance than that of CBP. This may be attributed to the adoption of the hosts, which have suitable HOMO/LUMO levels enabling excitons to form on host and hence favoring efficient energy-transfer from host to guest. In addition, the high bipolar carrier mobility of the host is found to be critical to this kind of doping system, which would balance the injection of both carriers and improve efficiency.

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1. Introduction

Organic light-emitting diodes (OLEDs) possess high potential in the development of full-color flat-panel displays and lighting [1–6]. In this regard, fabrication of OLEDs with higher power efficiency becomes essential. Phosphorescent OLEDs have attracted considerable attention because they can harvest both singlet and triplet excitons and achieve, in theory, 100% internal quantum efficiency, corresponding to a fourfold improvement in efficiency relative to that achievable in single-harvesting fluorescent OLEDs [7–13]. In general phosphorescence emitters are doped into a host [14–16]. And the dopant molecules usually get excited through direct charge trapping or energy transfer from the host [17,18]. However, for most of the phosphorescent OLEDs, the device quantum efficiency drops rapidly with increasing current density. This is believed to be attributed to the fact that triplet excitons relax more slowly and the emission inevitably reaches saturation through a quenching mechanism involving triplet-triplet annihilation [19]. In order to alleviate this problem, the host materials are significant to possess high bipolar carrier mobility and suitable highest occupied

* Corresponding author. Tel.: +86 5386715622; fax: +86 5386715916. *E-mail address:* xiaojingzx@163.com (J. Xiao).

http://dx.doi.org/10.1016/j.synthmet.2014.04.001 0379-6779/© 2014 Elsevier B.V. All rights reserved. molecular orbital (HOMO)/lowest unoccupied molecular orbital (LUMO) levels that match the neighboring layers [20,21]. To achieve this goal, tremendous effort has been devoted. In general, the bipolar host materials possess electron-transporting groups (electron acceptors) and hole-transporting units (electron donors) [22]. The donor-acceptor interaction of bipolar host materials is very critical, which have an influence on triplet energies. In addition, the HOMO-LUMO energy band gap (E_g) and triplet level (E_T) of the host materials should be higher than those of phosphorescent dopants, which can prevent reverse energy transfer from the guest back to the host. Nevertheless, a host material with a high E_T often has a wide E_g as a result of its typically large singlet-triplet energy difference (ΔE_{ST}) [23]. The host material with large E_g usually leads to direct charge trapping within the emitting layer (EML). This will produce poor conductivity in host material and high operation voltages in phosphorescent devices as result [24]. Host materials with narrow E_g are reported to have better performances [25–27]. In this case, design of bipolar host material with high E_T and narrow E_g is significant.

In this study, we report two novel kinds of 1,3,5-triazine derivatives, 3-(4,6-diphenyl-1,3,5-triazin-2-yl)-9-phenyl-9H-carbazole (DPTPCz) and 3-(4,6-diphenoxy-1,3,5-triazin-2-yl)-9-phenyl-9Hcarbazole (DPOTPCz), that contain 1,3,5-triazine as the electronaccepting core and carbazole-containing electron acceptors. The









Fig. 1. The molecular structures of DPTPCz and DPOTPCz.

two compounds were used as hosts for green and red iridium complex guests for highly efficient OLEDs. Since Baldo et al. developed the concept of phosphorescent OLEDs [28], 4,4'-N,N'-dicarbazolebiphenyl (CBP) has been used widely as a host material for phosphorescent OLEDs because of its suitable E_T and good hole-transporting ability [29–32]. We compared the electroluminescent (EL) properties of OLEDs based on the two 1,3,5-triazine derivatives with the devices using CBP as the host. We also investigated the effect of structural change on the performance of host:dopant system.

2. Experimental

New bipolar host materials DPTPCz and DPOTPCz have been synthesized in OLEDs. The molecule structures of these are shown in Fig. 1. To investigate the energy levels alignment of the two new materials, ultraviolet photoemission spectroscopy (UPS) of the two film samples was measured in a Kratos AXIS Ultra DLD ultrahigh vacuum surface analysis system. UPS analysis was performed with an unfiltered He I (21.2 eV) gas discharge lamp and a total instrumental energy resolution of 100 meV. The Fermi level (E_F) is referred to as the zero binding energy (BE) in the UPS spectra. In order to determine the LUMO positions of two 1,3,5-triazine derivatives, the film UV-vis absorption spectra were performed with Lambda 750, PerkinElmer. The excitation wavelength was scanned from 800 to 250 nm. All spectra were measured at room temperature.

For fabricating the devices, the patterned indium tin oxide (ITO, $6 \Omega/sq$) substrates (Lum Tech), organic (Nichem Fine Tech) and inorganic materials (Alfa Aesar), were commercially purchased. After the routine cleaning and ultraviolet (UV) ozone treatment, the ITO substrates were introduced into a highvacuum deposition chamber (Trovato MFG, base pressure about $1\times 10^{-6}\,\text{Torr})$ with multiple thermal evaporation sources, where film thickness was monitored in situ with a calibrated guartz crystal microbalance. N,N'-diphenyl-N,N'-bis(1-naphthyl)-(1,1'biphenyl)-4,4'-diamine (NPB), 4,4',4''-tris(N-carbazolyl)-triphenyl amine (TCTA), host-dopant composition, 1,3,5-tris(phenyl-2benzimidazolyl)-benzene (TPBi), lithium fluoride (LiF), and aluminum (Al) were evaporated on the substrates in turn by vacuum deposition. Phosphorescent bis(2-phenylpyridine)iridium acetylacetonate [Ir(ppy)₂(acac)] and tris(2-phenylquinoline) iridium(III) [Ir(2-phq)₃] were used as dopants for green and red emissions, respectively. Two sets of devices were fabricated in our experiment. The device architectures of set 1 and 2 are listed in Table 1. The comparative devices were fabricated in a same round under identical experimental conditions. The EL spectra and the current density-voltage-luminance (I-V-L) characteristics of the corresponding devices were measured simultaneously with a Photo Research PR-655 spectrometer and a computer controlled programmable Keithley model 2400 power source. All measurements were carried out at room temperature under ambient conditions after the devices have been encapsulated in a glove box.

Table 1

Layer structures of OLEDs GA, GB, GC, RA, RB and RC, where G-EML and R-EML refer to green and red electroluminescence units, respectively.

Devices or units	Layer structures	
Devices or units Device GA Device GB Device GC Device RA Device RB Device RC G-EML1 G-EML2	Layer structures ITO/NPB/TCTA/G-EML1/TPBi/LiF/AI ITO/NPB/TCTA/G-EML2/TPBi/LiF/AI ITO/NPB/TCTA/G-EML3/TPBi/LiF/AI ITO/NPB/TCTA/R-EML1/TPBi/LiF/AI ITO/NPB/TCTA/R-EML3/TPBi/LiF/AI ITO/NPB/TCTA/R-EML3/TPBi/LiF/AI DPTPC2:Ir(ppy) ₂ (acac)(7%; 30 nm)	
G-EML3	CBP:Ir(ppy) ₂ (acac)(7%; 30 nm)	
R-EML1	DPTPCz: $Ir(2-phq)_3(6\%; 30 nm)$	
R-EML2	DPOTPCz:Ir(2-phq) ₃ (6%; 30 nm)	
R-EML3	CBP:Ir(2-phq) ₃ (6%; 30 nm)	

3. Results and discussion

To understand the EL properties of $Ir(ppy)_2(acac)$ and $Ir(2-phq)_3$ doped in the 1,3,5-triazine derivatives hosts, we fabricated green and red OLEDs using DPTPCz and DPOTPCz as host materials with the configuration of ITO/NPB (30 nm)/TCTA (10 nm)/G-EML (or R-EML) (30 nm)/TPBi (30 nm)/LiF (1 nm)/Al (100 nm), where G-EML and R-EML refer to the 1,3,5-triazine derivatives layer doped with Ir(ppy)₂(acac) and Ir(2-phq)₃, respectively. The optimized doping concentration of Ir(ppy)₂(acac) is 7 wt.%, and 6 wt.% for Ir(2-phq)₃. For comparison, we also fabricated reference devices employing CBP as the host with the same configuration. The EL performances of those devices are summarized in Table 2. Device GA exhibits the highest lumen efficiency of 65.7 cd/A at 2.5 mA/cm², a maximum brightness of 50380 cd/m^2 at $7.13 \text{ V} (0-100 \text{ mA/cm}^2)$. And device GB exhibits the highest lumen efficiency of 64.8 cd/A at 2.5 mA/cm², a maximum brightness of 49740 cd/m^2 at $6.75 \text{ V} (0-100 \text{ mA/cm}^2)$. This phenomenon can also be observed when comparing the lumen efficiency of devices RA-RC. Device RA displays the highest lumen efficiency of 31.66 cd/A at 5 mA/cm², a maximum brightness of 22480 cd/m² at 6.43 V ($0-100 \text{ mA/cm}^2$), and device RB displays the highest lumen efficiency of 33.16 cd/A at 5 mA/cm², a maximum brightness of 26270 cd/m^2 at $6.63 \text{ V} (0-100 \text{ mA/cm}^2)$. It is

Table 2

Device performance parameters at current density of 10 mA/cm² and maximum luminance and luminous efficiency of green devices GA-GC, and red devices RA-RC.

Device	Voltage (V)	Luminous efficiency (cd/A)	Maximum luminance (cd/m ²)	Maximum luminous efficiency (cd/A)
GA	5.66	63.70	50,380	65.70
GB	5.11	62.93	49,740	64.80
GC	6.17	50.33	40,810	50.33
RA	4.96	31.22	22,480	31.66
RB	4.95	32.99	26,270	33.16
RC	6.15	24.35	18,520	25.80

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