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Optimum Structure Adjustment for Flexible Fluorescent and Phosphorescent Organic Light Emitting Diodes

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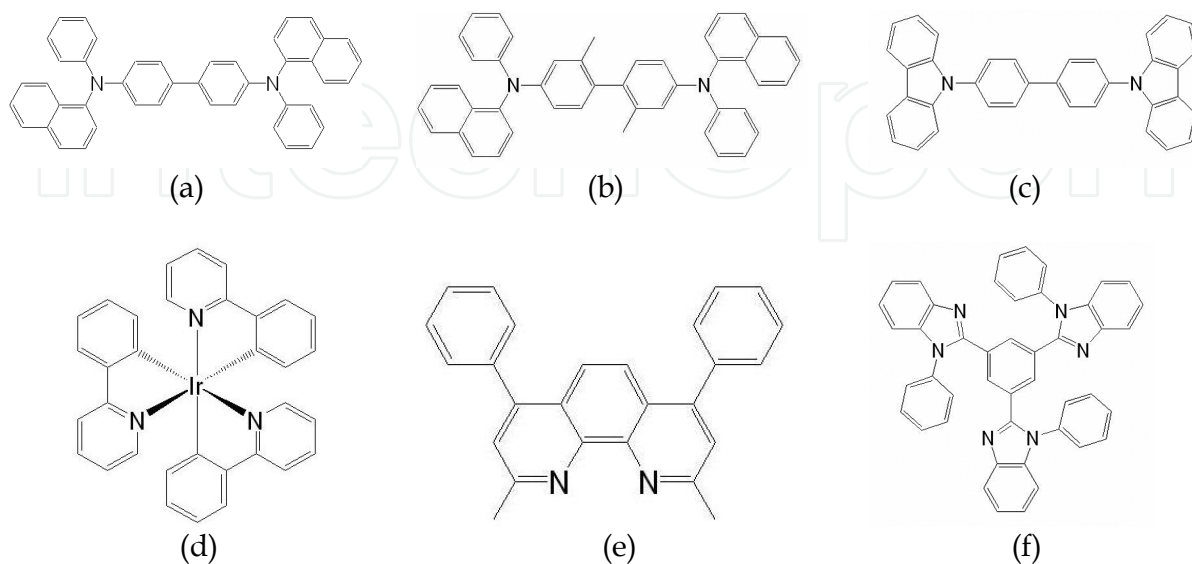
1. Introduction

The organic light emitting diodes (OLEDs) [1] is a new-generation flat panel display with the advantages of self-luminescence, wide viewing angle ($> 160^\circ$), prompt response time ($\sim 1 \mu\text{s}$), low operating voltage (3~10 V), high luminance efficiency, high color purity, and easy to be made on various substrates. Therefore, it's an important topic that how to improve the luminance efficiency, lifetime and the adhesion characters of ITO/organic interface of flexible OLEDs. Zungang Liu *et al.* reported that the NPB (HTL) is suitable in contact with the emission layer and when they form an energy ladder structure, the driving voltage decreased and the electroluminescent output increased [2]. Thus it can be seen, the hole transport layer [3-6] is very important to balance the injection of hole and electron, to increase the luminance efficiency and lifetime. In recent years, the hole buffer layer of device typically employs LiF [7], CuPc [8], Pani:PSS [9-10] or PEDOT:PSS [9-11] to improve the hole injection efficiency. In addition, a flexible substrate (PET, metal foil, etc.) surface is not completely smooth and will usually have spikes. After the organic thin film evaporates onto the ITO substrate surface the spikes will still exist. When the device is operated under high voltage or high current density, a heavy amount of electric current will concentrate at the spikes and damage the device by causing the device to short circuit, creating Joule heat. The luminance efficiency of the device will therefore be reduced producing shorter device lifetime. Thus, the PEDOT:PSS fabrication process uses spin-coating to obtain a thin film with a smoother surface than that produced by thermal deposition. Spin-coating enhances the organic material adhesion in subsequent processes, thereby directly affecting the performance of flexible OLED. For the above reason, this research dissolved hole transport material N,N'-diphenyl-N,N'-bis(1-naphthyl)-1,1'-biphenyl-4,4''-diamine (α -NPD), N,N'-Bis(naphthalene-1-yl)-N,N'-bis(phenyl)-benzidine (NPB) or α -NPD:NPB in tetrahydrofuran (THF) solvent and spin-coated the buffer layer onto ITO surface of flexible OLEDs. Phosphorescent dye gains energy from the radiative recombination of both singlet and triplet excitons [12], improving the internal quantum efficiency of fluorescent OLEDs (FOLEDs) typically 25% at maximum to nearly 100% [13]. Enhancing the luminance

efficiency of phosphorescent OLED has attracted the interest of many researchers. Improving device efficiency, the triplet state excitons must be confined in the emitting layer to increase the chance for energy transfer from host to guest. The material that achieves this effect is called the hole blocking layer (HBL), CF-X [14], CF-Y [14], BCP [12], TPBi [15], and BAlq [16]. These materials have higher ionization energy and band gap that can block the diffusion of excitons. When the host-guest orbit overlap is weak, the blocking layer action is particularly important.

2. Experiment

The ITO substrate used in this study was $80\Omega/\square$ PET substrate. Before depositing the patterned ITO substrate was placed in O_2 plasma for surface cleaning. The spin-coating solvents were then prepared by dissolving hole transport materials *N,N'*-diphenyl-*N,N'*-bis(1-naphthyl)-1,1'-biphenyl-4,4''-diamine (α -NPD) and *N,N'*-Bis(naphthalene-1-yl)-*N,N'*-bis(phenyl)-benzidine (NPB) (α -NPD mixed NPB with 1:1 wt%) in tetrahydrofuran (THF) solvent. The chemicals are vibrated ultrasonically in solution for 60 minutes to facilitate the dissolving process. The coating process is then carried out for 35 seconds at 4500 r.p.m. to deposit the buffer layer onto the ITO surface. After that the substrate was placed in an organic evaporation chamber to deposit the organic layers under 2×10^{-6} torr, α -NPD or NPB was deposited as hole transport layer (HTL), 4,4'-Bis(carbazol-9-yl) biphenyl (CBP) was deposited as the phosphorescent device host, Tris(2-phenyl-1-pyridine) iridium(III) ($Ir(ppy)_3$) was deposited as the phosphorescent device guest material, 2,9-Dimethyl-4,7-diphenyl-1,10-phenanthroline (BCP) or 2,2',2''-(1,3,5-Benzinetriyl)-tris(1-phenyl-1-H-benzimidazole) (TPBi) was deposited as the hole blocking layer (HBL), Tris(8-hydroxyquinolinato)aluminum (Alq3) was deposited as emitting layer (EML) of fluorescent and electron transport layer (ETL), and 1,3-Bis[2-(2,2'-bipyridine-6-yl)-1,3,4-oxadiazole-5-yl]benzene (Bpy-OXD) was deposited as electron transport layer/ hole blocking layer. The chemical structures of all used organic materials are shown in Fig. 1. SpectraScan PR650 and Keithley 2400 equipment were employed to measure the luminance and current-voltage characteristics.



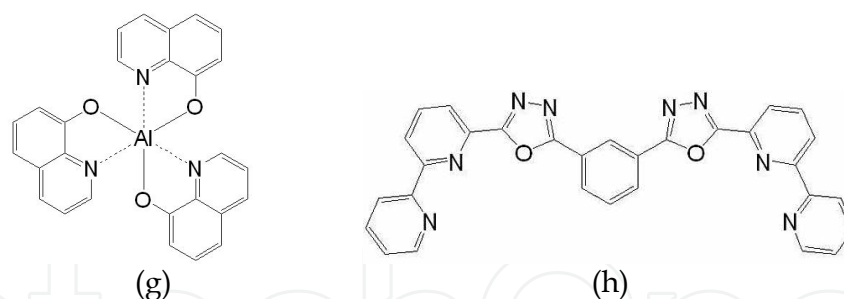


Fig. 1. The chemical structures of all used organic materials (a) NPB, (b) α -NPD, (c) CBP, (d) Ir(ppy)₃, (e) BCP, (f) TPBi, (g) Alq3 and (h) Bpy-OXD

3. Results and discussion

3.1 Optimum Structure Adjustment for Flexible Phosphorescent Organic Light Emitting Diodes

NO.	Sub.	ITO	NPB	Dopant 7% Ir(ppy) ₃ in CBP	HBL	Alq3	LiFAI	
A1	Plastic (PET)	ITO (80Ω/□)	40	20	BCP	40	0.5	
A2					0			
A3					5			
A4					10*			
B1			30	20	BCP	40		
A3			40					
B2			50*					
B3			70					
C1			50	20	BCP	30		50
B2						40		
C2						10		
C3						50*		
D1			50	10	BCP	10		50
C2				20				
D2				30				
D3	40*							
E1	50	40	TPBi	5	50			
E2				10*				
E3				15				

*optimum parameters

Table 1. Adjustment parameters of Phosphorescent organic light emitting diodes (unit: nm)

In this study the device structures are shown in Table 1. First, we inserted a hole blocking layer (HBL) to effectively confine the holes in the emitting layer (EML) for improving the luminance efficiency of the devices. Moreover, varied the thickness of BCP from 0 to 15 nm; it was found that the best hole-blocking result was present at 10 nm of the thickness of BCP (as shown in Fig.1). However, if the thickness of BCP was increased to 15 nm, the hole blocking result was better, but the distance of injecting electrons to EML was increased and caused the brightness decreased. Then, we tried to vary the thickness of NPB to make the amount of the hole injected into EML match with the amount of the electron for increasing the luminance efficiency of the device. From Fig. 2, it was found that the maximum luminance efficiency of the device can be obtained at 50 nm of the thickness of NPB. Furthermore, we varied the thickness of Alq3 to make the amount of the electron injected into EML match with the amount of the hole. From Fig. 3, it was found that the maximum luminance efficiency of the device can be obtained at 50 nm of the thickness of Alq3. However, if the thickness of Alq3 was increased to 70 nm, the distance of the electron injecting to EML was enhanced to decrease the amount of the electron injected into the EML to cause the brightness greatly decreased. At last, we varied the thickness of the EML of CBP:Ir(ppy)3 from 10 nm to 40 nm and hoped that the chance of recombining electron-hole will be increased via varying the thickness of the EML to increase the brightness and luminance efficiency. From the experiment result, it was found that the best luminance efficiency would be obtained at the layer thickness of 40 nm (as shown in Fig. 4); at the moment, the device efficiency was greatly increased to 30.4 cd/A.

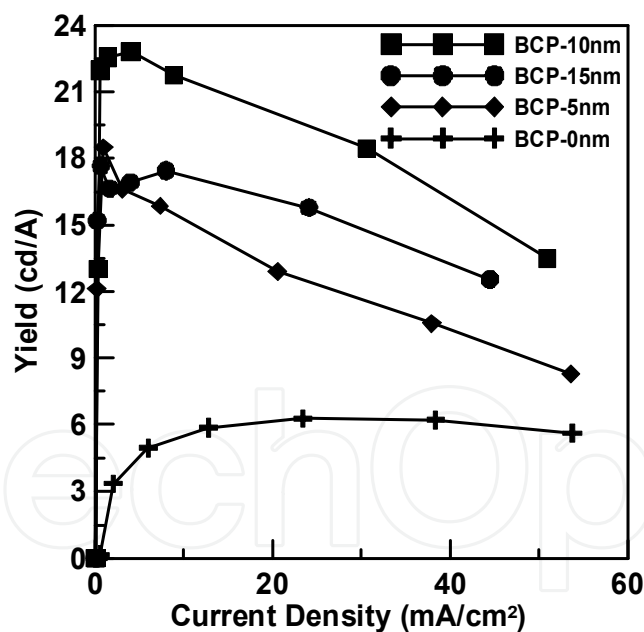


Fig. 1. Luminance efficiency-current density curves for different thicknesses of HBL

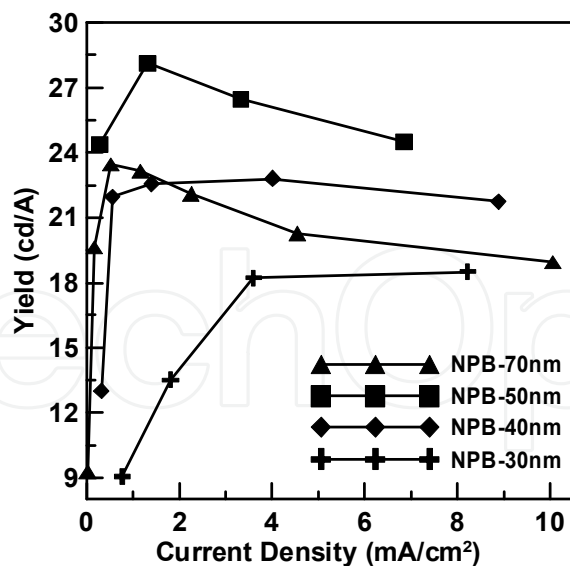


Fig. 2. Luminance efficiency-current density curves for different thicknesses of HTL

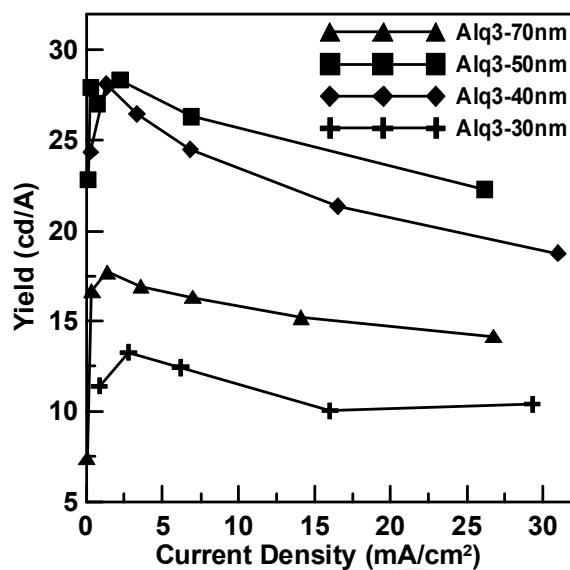


Fig. 3. Luminance efficiency-current density curves for different thicknesses of ETL

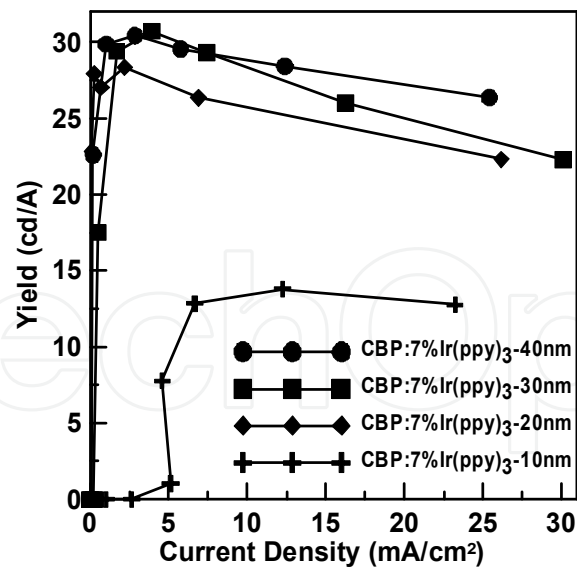


Fig. 4. Luminance efficiency-current density curves for different thicknesses of EML

Finally the doping concentration of $\text{Ir}(\text{ppy})_3$ was adjusted as shown in Table 2. We found in Fig. 5 that when the doping concentration was 7 wt% the device has optimum luminance efficiency. In this study we also changed different hole blocking layers (HBL) BCP and TPBi, whose structures are shown in Table 1, respectively. It is found from luminance efficiency - current density in Fig. 5 that using TPBi as hole blocking layer could enhance the luminance efficiency further to 34.2 cd/A. This is because the HOMO of TPBi (6.7 eV) is higher than the HOMO of BCP (6.3 eV), which confines holes and excitons in the emitting layer more effectively and prevents direct diffusion of holes or excitons to Alq₃ layer. After comparing the effects of different HBLs on luminance efficiency as shown in Fig. 5, it is found a suitable material of HBL can improve the luminance efficiency of device effectively.

From the results of these serial experiments, it is found that whenever the thickness of an organic layer has been adjusted, the luminance efficiency of the device has increased (as shown in Fig. 6). It is because the amount of electron-hole of the device becomes more balanced via the optimization of the hole transport layer and the electron transport layer. In addition, extending the charge carrier recombining area and using more suitable HBL material and suitable thickness of its thin film will gradually increase and improve the luminance efficiency of the device.

NO.	Sub.	ITO	NPB	Ir(ppy) ₃ :CBP		HBL	Alq ₃	LiF	Al
				concent	thickness				
F1	Plastic (PET)	ITO (80Ω/□)	50	4%	40	BCP 10	50	0.5	65
D3				7%*					
F2				8%					
F3				10%					

*optimum parameters

Table 2. Adjustment parameters of PHOLEDs with different $\text{Ir}(\text{ppy})_3$ doping concentrations (unit: nm)

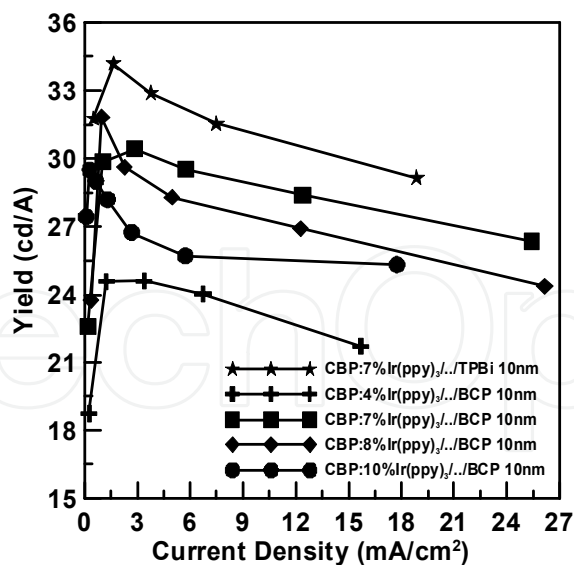


Fig. 5. Luminance efficiency-current density curves of PHOLEDs with different Ir(ppy)_3 doping concentrations and HBL materials

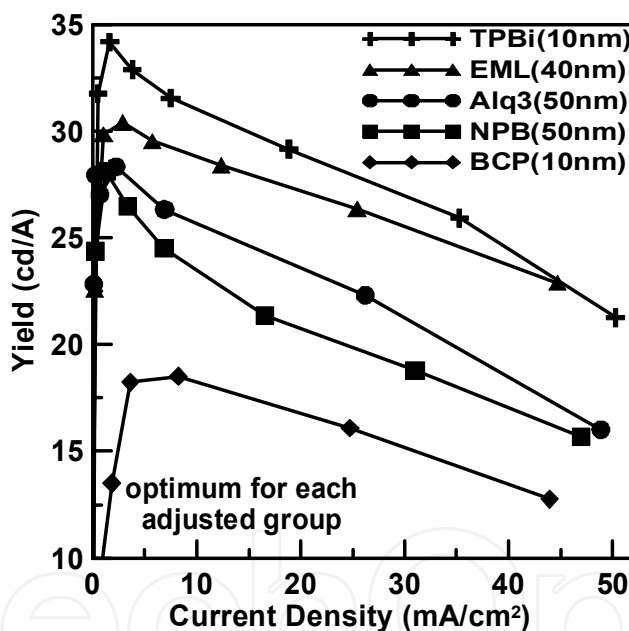


Fig. 6. Luminance efficiency-current density of optimum thickness for each adjusted layer

3.2 Spin-coating (α -NPD:NPB)+THF as the buffer layer and evaporating NPB as the hole transport layer to improve fluorescent OLED characteristics

Because the spin-coating rotation speed directly affects the film thickness, we fixed the spin-coating time at 35 seconds from the beginning of the experiment. The rotation speed of the spin- α -NPD:NPB layer (3500, 4000, 4500 and 5000 r.p.m.) was varied to study how the device optoelectronic characteristic was affected by the rotation speed of spin-coating. The device structure was PET/ITO (160 nm)/spin-coated (NPB: α -NPD)+THF (different rotation speed)/evaporated NPB (41 nm)/Alq3 (52 nm)/Bpy-OXD (15 nm)/LiF (0.5 nm)/Al (135 nm). The device energy band structure was shown in Figs. 7(a). Because the coated film

thickness will become thick under lower rotation speed, from Fig. 7(b), we can see the lowest current density flowing through the device at the lowest rotation speed (3500 r.p.m.). According to the Mott-Gurney rule [17], we know that the current density presents an inverse-proportion relation with the thickness, and the greatest NPB spin-coating layer thickness can be estimated to appear at a rotation speed of 3500 r.p.m. However, the layer thickness will become thinner as the rotation speed increases and the current density will increase accordingly. However, at 5000 r.p.m., we found that the device current density decreased. The major reason results from too high rotation speed which causes the centrifugal force to become too strong, preventing the NPB+THF solution from attaching onto the ITO surface; hence, the film thickness becomes uneven. From Fig. 8 luminance-current density characteristic curve, we can observe the same trend in the current density-voltage curve. Hence, we determined that 4500 r.p.m. is the optimal spin-coating parameter to get the best optoelectronic characteristic.

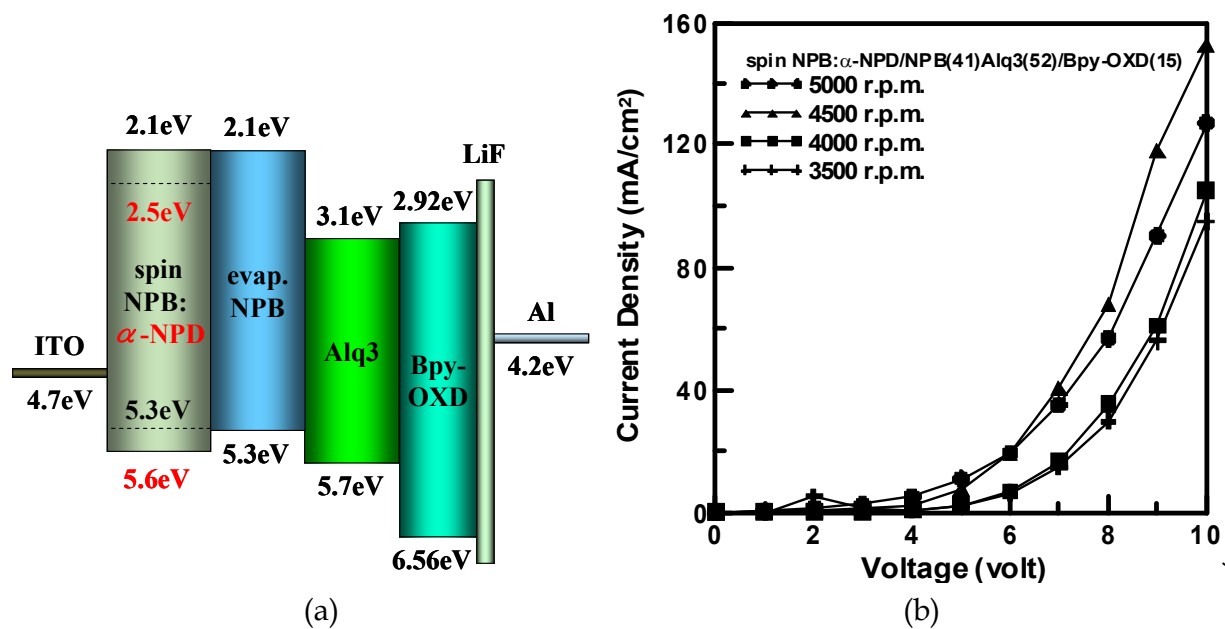


Fig. 7. (a) The energy band structure of the device with spin-coating (NPB: α -NPD)+THF buffer layer; (b) J-V characteristics for different rotation speeds

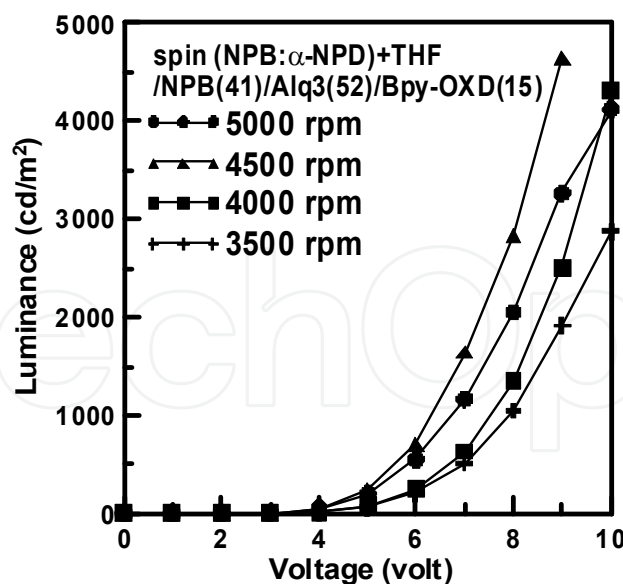


Fig. 8. L-V characteristics for spin-coating (NPB:α-NPD)+THF with different rotation speeds

To further verify the first HTL (NPB:α-NPD)+THF manufacturing process versus improving the device optoelectronic characteristics, the following experiment was conducted. Devices with PET/ITO(160 nm)/ HTL/Alq3(52 nm)/Bpy-OXD(15 nm)/LiF(0.5 nm)/Al(135 nm) structures wherein the HTL was produced via the spin-coating or thermal-evaporation process or both, are shown in Table 3.

Under same current density, the optoelectronic characteristics of Devices G1 and G2 (employing both spin-coated and evaporated double HTLs structures) were higher than that of traditional devices made with single HTL using only thermal-evaporation (Device G3), as shown in Fig. 9(a) and 9(b). The device characteristics with double HTLs were also found better than those with the single HTL spin-coated structure (NPB:α-NPD)+THF only (Device G4). Because spin-coating the film in the amorphous mode on the ITO surface does not easily generate pin holes, and the organic layer contact with the ITO becomes tighter, the HTL surface roughness can be greatly improved [18-21]. Therefore, the holes are injected more easily from the ITO electrode into the NPB layer. Furthermore, the optoelectronic characteristics of Device G1 were better than Device G2 because spin-coating (NPB:α-NPD)+THF has similar p-type doping properties that enable holes to be injected efficiently [22]. NPB and α-NPD co-doping will produce band bending and enable the holes to tunnel from the ITO electrode into the HTL, forming an Ohmic contact interface [23]. Consequently the Device G1 exhibited the best performances than the other devices. Fourier Transform Infrared Spectroscopy (FTIR) was also performed on the HTL films to study the variation in molecular structure. Moreover, from the lifetime test results shown in Fig. 10, the lifetime of Device G1 was the longest, increased by about 41 % longer than Device G3 (fabricated without spin-coating (NPB:α-NPD)+THF layer).

Devices	HTL thickness	
	(NPB: α -NPD) dissolved in THF then spin-coating	evaporate
G1	spin (NPB: α -NPD)+THF	NPB
	58 nm	41 nm
G2	spin NPB +THF	NPB
	37 nm	41 nm
G3	spin (NPB: α -NPD)+THF	NPB
	0 nm	41 nm
G4	spin (NPB: α -NPD)+THF	NPB
	58 nm	0 nm

Table 3. Different parameters of hole transport layer structures (unit: nm)

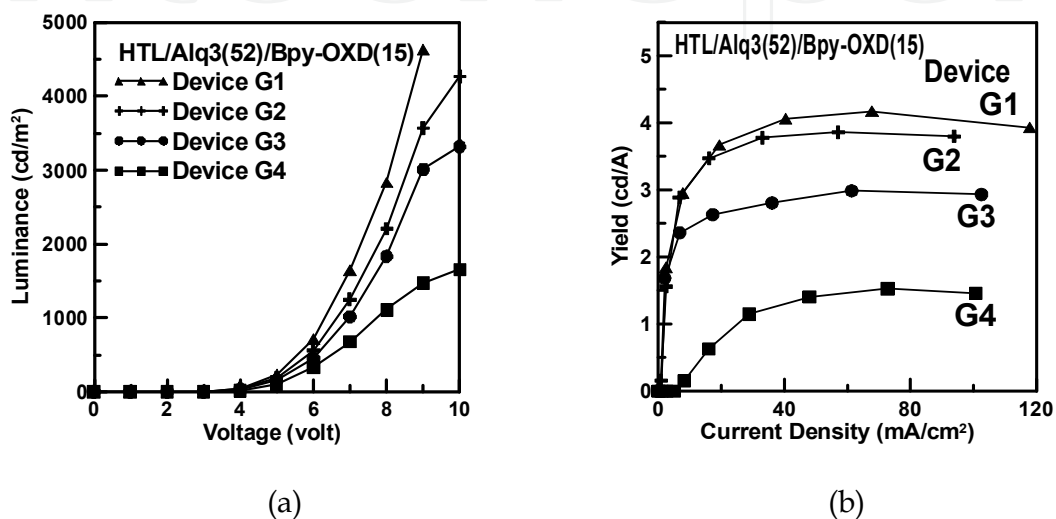


Fig. 9. (a) L-V and (b) Y-J characteristics for different HTL structures

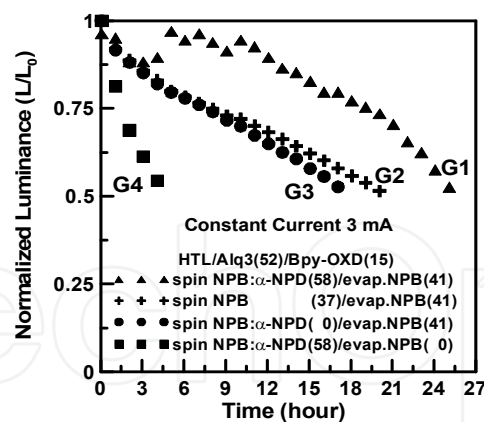


Fig. 10. The lifetime of flexible OLEDs with different HTL structures, and the devices were packaged by evaporating m-MTDATA 500 nm on the device surface

4. Conclusion

This research successfully improved the luminance efficiency and lifetime of flexible fluorescent and phosphorescent organic light emitting diodes by optimizing organic layer thicknesses or inserting a spin-coated buffer layer. From the results, the best phosphorescent device structure (ITO/ NPB (50nm)/ Ir(ppy)₃:CBP (40nm)/ TPBi (10nm)/ Alq3 (50nm)/ LiF

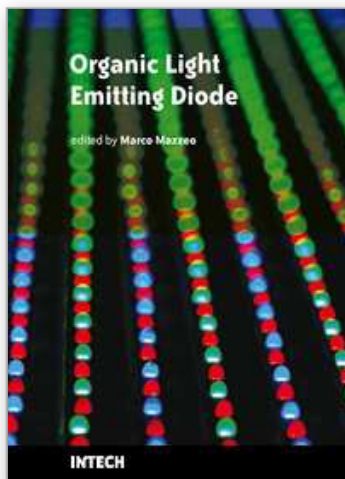
(0.5nm)/ Al (65nm)) has a maximum luminance efficiency of 34.2 cd/A by optimizing organic layer thicknesses. This device performance improvement is attributed to increased hole injection and improved hole-electron balance. In addition, extending the charge carrier recombining area and using more suitable HBL material and suitable thickness of its thin film will gradually increase and improve the luminance efficiency of the device. Comparing devices with TPBi and BCP as the HBL, it was found that devices fabricated with TPBi as the HBL exhibited better luminance efficiency than BCP.

We also discussed the effect with/without a spin-coated buffer layer on device performance. It was found that devices inserted with a spin-coated buffer layer had better brightness, luminance efficiency and lifetime than conventional devices (without spin-coating buffer layer). Furthermore, the research demonstrated the advantage of using spin-coating co-doping (NPB: α -NPD)+THF (exhibit the similar p-type doping properties and metal-like Ohmic contact interface) plus thermal-evaporation in fabricating flexible fluorescent organic light emitting diodes with double-hole transport layer structures. The performance of the best Device G1 exhibited a luminance of 4634 cd/m² at 9 V and a luminance efficiency 4.18 cd/A at 68 mA/cm². Compared with the single thermal-evaporated NPB only HTL structure, the luminance efficiency of device with spin-coated (NPB: α -NPD)+THF as buffer layer was increased by about 1.32 cd/A and its half-lifetime was increased by about 41 % longer than the device without spin-coated buffer layer.

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Organic Light Emitting Diode

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Organic light emitting diodes (OLEDs) have attracted enormous attention in the recent years because of their potential for flat panel displays and solid state lighting. This potential lies in the amazing flexibility offered by the synthesis of new organic compounds and by low-cost fabrication techniques, making these devices very promising for the market. The idea that flexible devices will replace standard objects such as television screens and lighting sources opens, indeed, a new scenario, where the research is very exciting and multidisciplinary. The aim of the present book is to give a comprehensive and up-to-date collection of contributions from leading experts in OLEDs. The subjects cover fields ranging from molecular and nanomaterials, used to increase the efficiency of the devices, to new technological perspectives in the realization of structures for high contrast organic displays and low-cost organic white light sources. The volume therefore presents a wide survey on the status and relevant trends in OLEDs research, thus being of interest to anyone active in this field. In addition, the present volume could also be used as a state-of-the-art introduction for young scientists.

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