

Double Hole Transport Layers Deposited by Spin-coating and Thermal-evaporating for Flexible Organic Light Emitting Diodes

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Abstract

The research applied the processes of spin-coating and thermal-evaporating in proper order to deposit the hole transport material N,N'-Bis(naphthalen-1-yl)-N,N'-bis(phenyl)-benzidine (NPB) on the ITO substrate to make flexible organic light emitting diodes (FOLED) with double hole transport layer.

1. Introduction

The organic light emitting diodes (OLED)[1-2] is a new-generation flat panel display with the advantages of self-luminescence, wide viewing angle ($> 170^\circ$), 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. Normally, OLED is made on glass substrate via the process of thermal-evaporating, but being made on plastic substrate will make OLED become light, thin and flexible[3]. Hence, OLED which is being made on plastic substrate will be a future trend.

Since the surface of ITO substrate is not completely level and smooth and the pointed-end protrusion will still be existing after the completion of the process of thermal-evaporating has been made, when the device is operated under high voltage or high current density, considerable current density will appear at the pointed-end protrusion and will damage the device or cause the device short circuit and create Joule heat; this will decrease the luminance efficiency of the device and decay its lifetime. Therefore, this study applied an extra process of spin-coating hole transport layer to improve the level and smoothness of the surface of the substrate.

2. Experimental

The ITO substrate applied in the study was $35 \pm 5 \Omega/\square$ polyethylene terephthalate (PET) substrate. Before depositing the organic layer, ultrasonically cleaning the patterned ITO substrate in proper order with acetone (10 seconds), methanol (8 minutes) and ion exchange water (8 minutes); then, we dried it via blowing nitrogen and put it into oven for roasting at 90°C for 8 minutes. Then, put plastic substrate into 30 W O_2 Plasma cleaner to clean the surface of ITO for improving its work function. The experiment applied NPB as the hole transport material, wherein NPB will be deposited via the process of thermal-evaporating or spin-coating. The solution applied for spin-coating was NPB powder dissolved into the solvent of TETRAHYDRIFURAN (THF), and the concentration was 0.2 wt% (NPB+THF solution); then, we mixed it well with ultrasonic cleaning for 60 minutes to assure its complete dissolution, and after that, the solution was filtered with a $0.2 \mu\text{m}$ filter to remove impurities. In the atmosphere, after completing the spin-coating NPB+THF solution, we put test piece into the organic thermal-evaporating vacuum chamber and then proceeded the processes, in proper order, such as thermal-evaporating hole transport layer(NPB), light emitting layer (Tris(8-hydroxy-quinolinato) aluminum, Alq3), electron transport concurrently as hole blocking layer(1,3-Bis[2-(2,2'-bipyridine-6-yl)-1,3,4-oxadiazole-5-yl]benzene, Bpy-OXD)[4] and finally proceeded the deposition of LiF and Al cathode. The device luminous area was $0.5 \times 0.5 \text{ cm}^2$. After completing device preparation, optoelectronic character was measured in the atmosphere, and Keithley 2400 was applied to be the power source and current-voltage characteristics was measured; in addition, SpectraScan PR-650 was applied to measure the

luminance and spectra.

3. Results and discussion

Since the rotation speed of spin-coating will directly affect the thickness of the film, we fixed the spin-coating time at 35 seconds from the beginning of the experiment but varied the rotation speed of spin NPB layer (3500, 4000, 4500 and 5000 r.p.m.) to study how the device luminance characteristic was affected by the thickness of spin-coating. The structure of the device of the research was PET/ ITO (160 nm)/ spin NPB (various rotation speed)/ evaporate NPB (41 nm)/ Alq3 (52 nm)/ Bpy-OXD (15 nm) / LiF (0.5 nm)/ Al (135 nm). The energy band of the device as shown in Fig. 1, and Fig. 2 is the character curve of device current density versus voltage under various rotation speed of NPB+THF spin-coating.

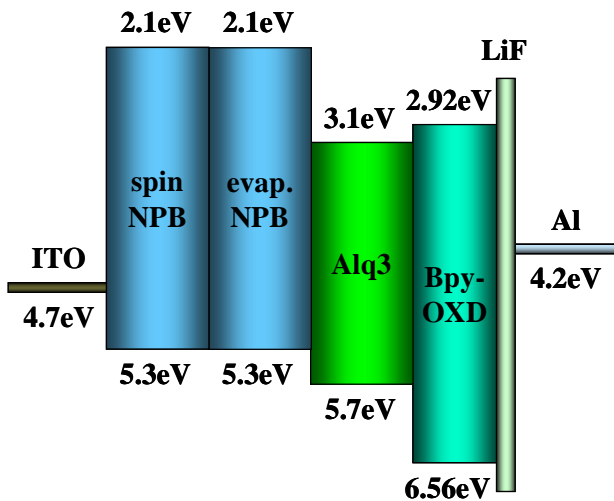


Fig. 1. The energy band of the device.

Since the thickness of the coated film will become thicker under lower rotation speed, from Fig. 2, 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[5], we know that current density presents an inverse-proportion relation with the thickness, and can estimate that the thickest thickness of the spin-coating NPB layer will appear at the rotation speed of 3500 r.p.m., but the thickness will become thinner while rotation speed goes faster, and current density will increase accordingly. However, at 5000 r.p.m., we found that

the device current density was decreased. The major reason could be resulted from a too fast rotation speed which causes centrifugal force to become too big to make the NPB+THF solution unable to attach on the ITO surface easily; hence, the film thickness becomes uneven. Fig. 3 shows the curve of luminance versus voltage.

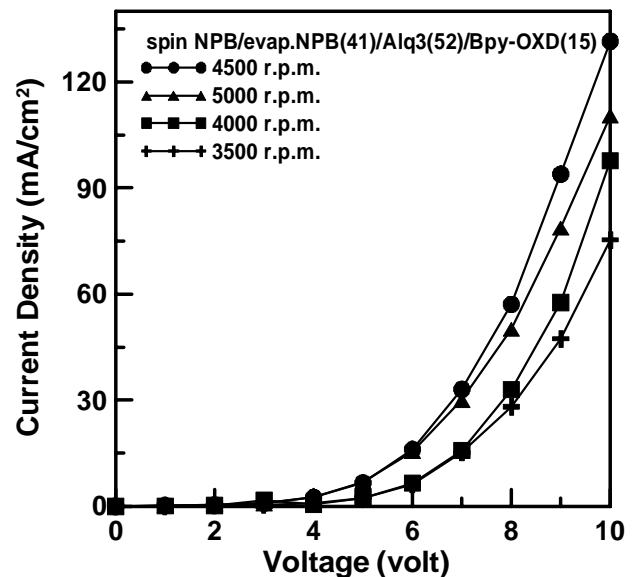


Fig. 2. Comparison of current density versus voltage under different rotation speed.

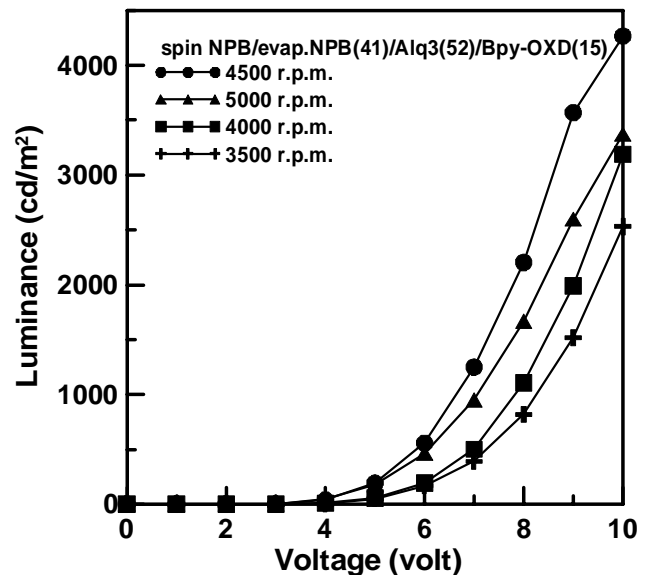


Fig. 3. Comparison of luminance versus voltage under different rotation speed.

From Fig. 3, we also can recognize the same trend as the curve of current density-voltage. Hence, we determine that NPB+THF 4500 r.p.m. is the best parameter of spin-coating to get the best luminance characteristic.

For further attesting the effect of improving the luminance characteristic of the device via the process of the hole transport layer NPB+THF spin-coating, we proceeded with the following experiment to study how spin-coating NPB layer affected the luminance characteristic of the device. The device structure was PET/ ITO (160 nm)/ HTL/ Alq3 (52 nm)/ Bpy-OXD (15 nm)/ LiF (0.5 nm)/ Al (135 nm); wherein HTL was the hole transport layer and can be made via the process of spin-coating or thermal-evaporating or both. As shown in TABLE 1, HTL are grouped into four, Device A spin NPB+THF (37 nm)/ evaporate NPB (41 nm), Device B evaporate NPB (78 nm) only, Device C evaporate NPB (41 nm) only, Device D spin NPB+THF (78 nm) only, wherein HTL total thickness of Devices A, B and D being maintained at the same.

TABLE 1. Different deposition methods for double HTLs in devices

Devices	HTL thickness	
	dissolved in THF then spin-coating	evaporate
A	spin NPB+THF 37 nm	NPB 41 nm
B	spin NPB +THF 0 nm	NPB 78 nm
C	spin NPB+THF 0 nm	NPB 41 nm
D	spin NPB+THF 78 nm	NPB 0 nm

Fig. 4 and 5 is the character curve of the relation of luminance versus current density and luminance efficiency versus current density respectively. From the figures, we can find that under same current density, comparing with conventional single thermal-evaporating NPB structure (Device B and C), spin-coating plus thermal-evaporating double NPB structure (Device A) has higher luminance and luminance efficiency, and, in addition, has better character than that of the device with single spin-coating NPB+THF structure (Device D). The reason is

that NPB film on ITO surface made via spin-coating is amorphous and not easy to create pin-hole; hence, NPB film will be contacted with ITO more tightly and ITO surface roughness will be improved considerably. Accordingly, the result will help the hole to inject NPB layer from ITO electrode. Moreover, NPB film made via thermal-evaporating above spin NPB is homojunction with spin NPB, and hole-transport is smooth too.

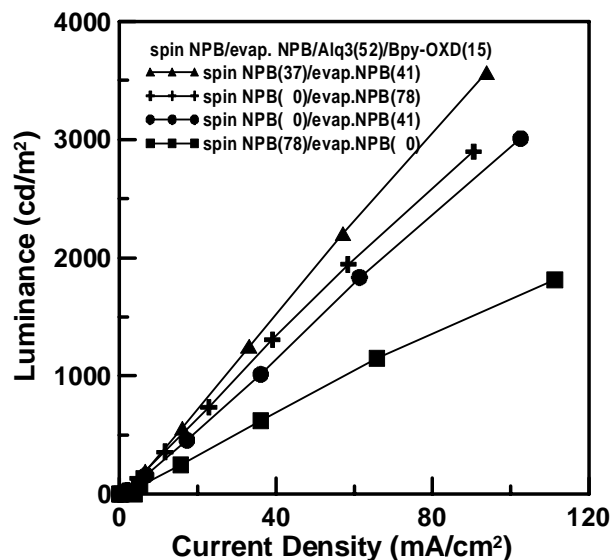


Fig. 4. Comparison of luminance versus current density under various hole transport layer.

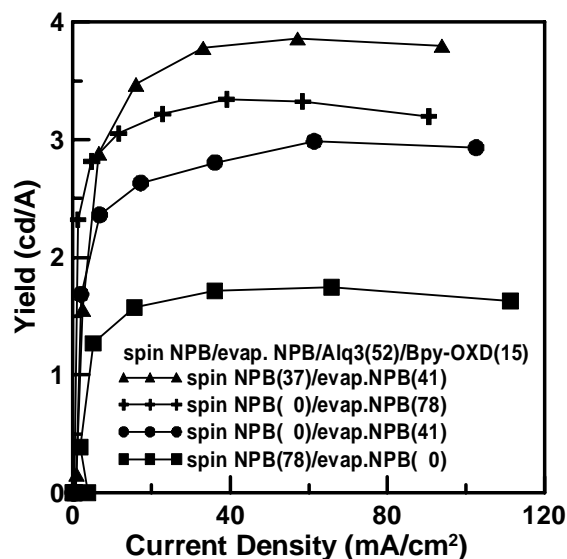


Fig. 5. Comparison of yield versus current density under various hole transport layer.

Therefore, the device has the best character. From above discussion, it shows that spin-coating can improve the ITO surface roughness, but, comparing Device A with Device D, we found that, at same HTL thickness, spin-coating plus thermal-evaporating double layers NPB structure had 2 cd/A with higher luminance efficiency than that of single layer spin-coating NPB+THF (78 nm) structure, as shown in Fig. 5 The best structured device has the maximum luminance 3568 cd/m² at 9 V and the maximum luminance efficiency 3.86 cd/A at 8 V. The Fourier Transform Infrared Spectroscopy (FTIR) was performed on the HTL films by spin-coating or evaporation to study the quality variation of molecular structure.

From the lifetime test results as shown in Fig. 6, the lifetime of Device A is the longest, it has been increased about 14.4 % longer than Device C which was fabricated without spin-coating NPB+THF layer.

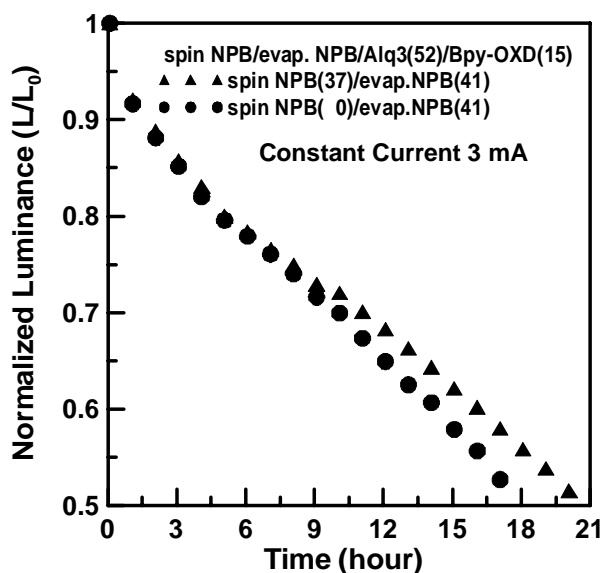


Fig. 6. The lifetime of OLEDs with various HTL structures, and the devices were passivated by evaporating m-MTDATA 500 nm on the device surface.

4. Summary

The study successfully applied the processes of spin-coating and thermal-evaporating in proper order to deposit the hole transport material (NPB) to make the flexible organic light emitting diodes of the double hole transport layers. When the device structure was PET/ ITO (160 nm)/ spin NPB (37 nm)/ evaporate NPB (41 nm)/ Alq3 (52 nm)/ Bpy-OXD (15 nm)/ LiF (0.5 nm)/ Al (135 nm), we got the maximum luminance 3568 cd/m² at 9 V and the maximum luminance efficiency 3.86 cd/A at 8 V; the luminance efficiency was increased 1 cd/A, comparing with those which are made without spin-coating hole transport.

5. References

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