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Co-doping in Spin-coated Hole Transport Layer for Flexible Organic Light Emitting Diodes

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1. Introduction

When using thermal-evaporating to deposit organic materials, the technique of co-doping is often employed to change emitting color and enhance luminance efficiency of organic light emitting diodes (OLED)[1]. But when the organic material is being used at a very small amount, the thermal-evaporating is usually unstable, resulting in the impossibility of accurate control of the doped-concentration. Therefore, when spin-coating is being used in this study, and two dopants (co-doping)[2] both are dissolved in the solution, the roughness of the substrate surface will be improved, and the co-doping ratio will be accurately adjusted, to further improve the electro-optical properties of OLEDs.

2. Experiment

The ITO substrate used in the study was 35±5 Ω/□ flexible polyethylene terephthalate (PET) substrate. In the device structure of this study, there existed double hole transport layers. The first hole transport layer (HTL) was fabricated by way of spin-coating, The solution applied for spin-coating is N,N'-Bis(naphthalen-1-yl)-N,N'-bis (phenyl) -benzidine (NPB) and N,N'-diphenyl- N,N'-bis(1-naphthyl) -1,1'biphenyl-4,4''diamine (α-NPD) powders dissolved into the solvent of TETRAHYDRFURAN (THF) in proper ratio (in this paper, it is abbreviated as (NPB:α-NPD)+ THF). Ultrasonic is employed to vibrate 60 minutes to assure their complete dissolution, and the concentration is 0.2 wt%. A filter of 0.2 μm is also used to remove the impurities in the solution. The second HTL was deposited by thermal evaporation. The emitting layer and electron transport layer is Tris(8-hydroxy-quinolinato) aluminum (Alq3) and 1,3-Bis[2-(2,2'-bipyridine-6-yl)-1,3,4-oxadiazo -5-yl] benzene (Bpy-OXD), respectively, also by evaporation. The device light-emitting area was 0.5×0.5 cm^2.

3. Results and Discussion

We fixed the spin-coating time at 35 seconds from the beginning of the experiment, varying the rotation speed of spin-coating (differentiate into 3500, 4000, 4500 and 5000 rpm) for the (NPB:α-NPD)+THF layer, in order to study the effect of spin-coating thickness on the luminance characteristic of the devices. The device's structures are PET/ ITO(160 nm)/spin (NPB:α-NPD)+THF (different 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 is shown in Fig. 1. The characteristic of the luminance versus voltage for different rotation speeds of spin-coating (NPB:α-NPD)+THF of the devices is shown in Fig. 2. As the film which is being coated at a low rotation speed is relatively thick, the luminance intensity of the device will become higher when the rotation speed is getting higher (4500 rpm) and the film is getting thinner. But when the rotation speed gets as high as 5000 rpm, the device luminance will be found to reduce contrarily, resulting from the reason that when the rotation speed is getting too fast, the centrifugal force is getting too strong. Consequently it leads to the uneasy adhesion of the solution of (NPB:α-NPD) +THF onto the surface of ITO. Therefore, acquirement of 4500 rpm is the best parameter of spin-coating to get the best luminance characteristic.

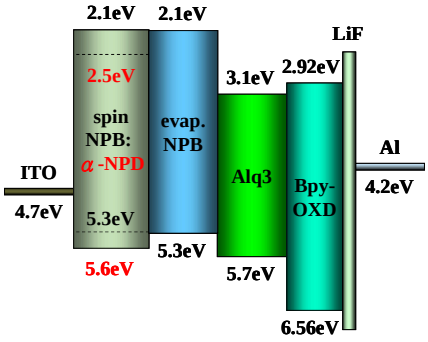

Fig. 1 The energy band of the device.

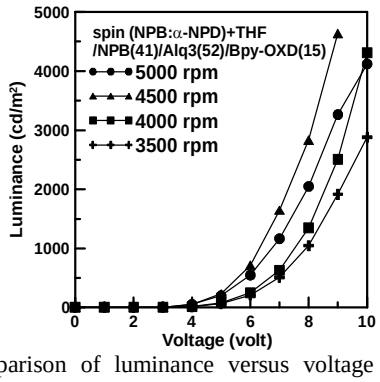

Fig. 2 Comparison of luminance versus voltage for different rotation speed.

In order to further prove the manufacturing process of spin-coating of the first HTL (NPB:α-NPD)+THF versus the effect of improving the luminance characteristic of devices, the following experiment is, therefore, proceeded. The devices, which have structures of PET/ITO(160 nm)/ HTL/Alq3(52 nm)/Bpy-OXD(15 nm)/LiF(0.5 nm)/Al(135 nm), wherein HTL can be made via the process of

spin-coating or thermal-evaporating or both, as shown in Table I.

Table I. Different deposition methods for double HTLs in devices

Devices	HTL thickness	
	dissolved in THF then spin-coating	evaporate
A	spin (NPB:α-NPD)+THF 58 nm	NPB 41 nm
B	spin NPB +THF 37 nm	NPB 41 nm
C	spin (NPB:α-NPD)+THF **0 nm**	NPB 41 nm
D	spin (NPB:α-NPD)+THF 58 nm	NPB **0 nm**

Fig. 3 and 4 are the characteristic curves indicating the relation of luminance versus voltage and luminance efficiency versus current density, respectively. Here it can be found that under same current density, the luminance efficiency of spin-coating plus thermal-evaporating double hole transport layer structures (Devices A and B) is higher than the traditional devices made with single HTL by thermal-evaporating only (Device C). The characteristic of the device with double hole transport layers are also found better than those with single HTL structure by spin-coating (NPB:α-NPD)+THF only （Device D）. Because by means of using spin-coating to make the film in amorphous mode on the surface of ITO pin-hole is not easy to be generated, and the organic layer contacted with ITO can become tighter and thus the surface roughness of HTL can be greatly improved. Therefore, it will help the holes to inject more easily from the electrode of ITO into NPB layer.

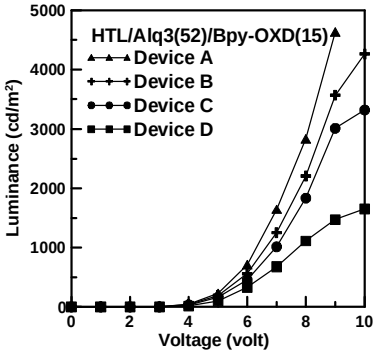

Fig. 3 Comparison of luminance versus voltage for various HTL structures.

The luminance and luminance efficiency of Device A are better than Device B, that is because spin-coating (NPB:α-NPD)+THF has similar p-type doping properties to enable holes to be injected efficiently[3], and NPB and α-NPD co-doping will make band bending and enable the holes by the way of tunnelling from the ITO electrode into the HTL and form an ohmic contact interface[4], consequently the characteristics of the device is deemed the best. The Fourier Transform Infrared Spectroscope (FTIR) was performed on the HTL films by spin-coating or evaporation to study the quality variation of molecular structure.

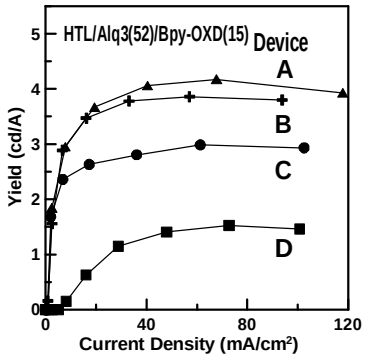

Fig. 4 Comparison of luminance efficiency versus current density for various HTL structures.

From the lifetime test results as shown in Fig. 5, the lifetime of Device A is the longest, it has been increased about 41 % longer than Device C which was fabricated without spin-coating (NPB:α-NPD)+THF layer.

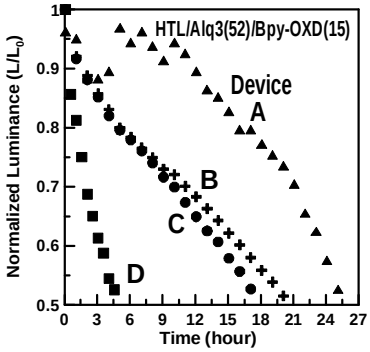

Fig. 5 The lifetime of OLEDs with various HTL structures, and the devices were passivated by evaporating m-MTDATA 500 nm on the device surface.

4. Conclusions

We have demonstrated the advantage of using spin-coating co-doping (NPB:α-NPD)+THF plus thermal-evaporating NPB technique in the case of fabricating flexible organic light emitting diodes with the double hole transport layer structures. The performance of the best device A exhibited a luminance 4634 cd/m² at 9 V and a luminance efficiency 4.18 cd/A at 68 mA/cm². Compared with the single thermal-evaporating NPB only as the structure of HTL, the luminance efficiency of device A will be increased about 1 cd/A and its half-lifetime will also increased about 41 % longer.

Acknowledgements

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