

Research paper

Achieving high efficiency by high temperature annealing of hole transporting polymer layer in solution-processed organic light-emitting devices



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ABSTRACT

We developed highly efficient solution-processed phosphorescent organic light-emitting device by annealing of hole transporting polymer layer at about glass transition temperature. Poly(*N,N'*-bis(4-butylphenyl)-*N,N'*-bis(phenyl)benzidine) (poly-TPD) was used as a hole transporting polymer layer and tris(2-phenylpyridine)iridium (III) [Ir(ppy)₃] doped *N,N'*-dicarbazolyl-3,5-benzene (mCP) was used as a solution-processed emission layer. The annealing temperature was critical to the well-defined interface between the poly-TPD and mCP:Ir(ppy)₃ layers. In addition, the carrier recombination was significantly enhanced by high temperature annealing of poly-TPD layer. A current efficiency of 61.5 cd/A and an external quantum efficiency of 17.5% were achieved by annealing of poly-TPD layer at about glass transition temperature.

1. Introduction

Since the first report on efficient bilayer organic light-emitting devices (OLEDs) by C. W. Tang, aggressive researches have been conducted during the past decades because they can provide superior characteristics such as fast response, wide viewing angle, and thin thickness in displays and lighting applications [1]. The external quantum efficiency (EQE) of OLEDs has been significantly improved by the phosphorescent organic materials that can harvest both the singlet and triplet excitons [2]. Red, green, and blue phosphorescent OLEDs with EQEs of 30–35% have been demonstrated by using Ir-complexes [3–6]. These high efficiency devices have been prepared by the vacuum evaporation process that can stack multiple organic layers for improving the carrier recombination, the radiative decay of excitons, and the extraction of light. However, there are only few reports on highly efficient solution-processed OLEDs although the solution processing can provide several advantages of low cost and large area fabrication. [7–9]. In realizing solution-processed OLEDs, the hole transport layer should have sufficient stability to the subsequent solution processing steps for the multilayer structures.

In this paper, we report highly efficient solution-processed OLEDs by annealing of hole transport layer at about glass transition temperature. Poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS) has been widely used as a hole injection or hole transport layer for the solution-processed OLEDs because of its high optical transparency in the visible

range, high electrical conductivity, and reasonable work function for hole injection [10–14]. PEDOT:PSS is soluble in water so that it provides a wide degree of freedom in selecting the solvents for solution coating of subsequent layers on the PEDOT:PSS layer. However, it has several limitations such as high acidity, inefficient blocking of electrons, and thermal instability [10]. Therefore, many researchers have investigated the alternatives for PEDOT:PSS. Poly(*N,N'*-bis(4-butylphenyl)-*N,N'*-bis(phenyl)benzidine) (poly-TPD) has been used as a general hole transport material for the solution-processed devices because it has a high hole mobility and reasonable highest occupied molecular orbital (HOMO) energy level to inject holes [15–18]. The poly-TPD is soluble in common organic solvents such as chlorobenzene, toluene so that the selection of solvents for subsequent layers can be limited. Therefore, the poly-TPD layer has been typically annealed at temperature less than 150 °C before further processing. However, the annealed poly-TPD can also be dissolved by the solvents for the subsequent layers [17]. In this paper, we report that the solvent resistance of poly-TPD layer can be significantly improved by annealing at about glass transition temperature. We also demonstrate the highly efficient solution-processed OLEDs by annealing of poly-TPD layer at about glass transition temperature.

2. Experimental

Indium tin oxide (ITO) coated glass substrates were used to fabricate the OLEDs. The sheet resistance of ITO film was about 10 Ω/sq. The ITO coated substrates were cleaned with acetone, methanol, and deionized

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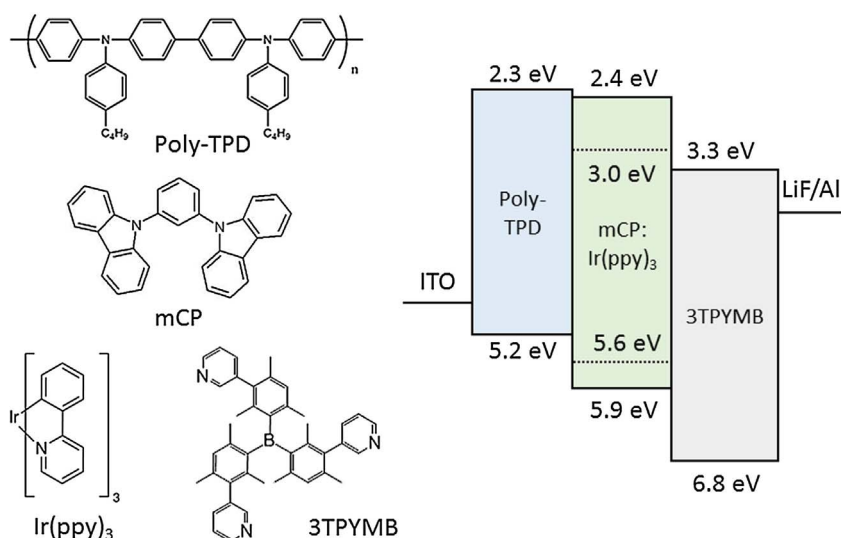


Fig. 1. Chemical structures of the organic materials and energy diagram of the device.

water after defining the anode patterns using a standard photolithography process. The ITO anode patterns were exposed to oxygen plasma for removing the residual organics and increasing the work function before spin-coating of the poly-TPD solution in chlorobenzene. The spin-coated poly-TPD layer was annealed at temperatures of 50, 150, and 220 °C for 30 min in air. After then, a 20 nm thick emission layer was spin-coated from tris(2-phenylpyridine)iridium(III) [Ir(ppy)₃] doped N,N'-dicarbazolyl-3,5-benzene (mCP) solution in chlorobenzene. After spin-coating of the mCP:Ir(ppy)₃ layer, a 50 nm thick tris[3-(3-pyridyl)-mesityl]borane (3TPYMB) layer was deposited by using a vacuum evaporation method at a base pressure of 1×10^{-6} Torr. After then, 0.5 nm thick LiF and a 100 nm thick Al layers were sequentially evaporated to define the cathodes through a shadow mask without breaking a vacuum. The completed structure was ITO/poly-TPD/mCP:Ir(ppy)₃/3TPYMB/LiF/Al. Fig. 1 shows the chemical structures of the organic materials and the energy diagram of the device. The active area of the device was 4×4 mm². The surface roughness, topology, and thickness of the organic layers were measured by atomic force microscopy (AFM, PSIA XE-100) and surface profiler (Kosaka Lab. ET3000), respectively. Current density-voltage-luminance characteristics of the devices were measured using computer controlled Keithley 2400 source-measured units and a calibrated Si photodiode. Electroluminescence (EL) spectra of the devices were measured with a Minolta CS1000 spectrophotometer. EQE of OLED was measured by using a luminance data, EL spectra, current density, eye-sensitivity curve [18].

3. Results and discussion

Chlorobenzene was used as a solvent for spin-coating of the mCP:Ir(ppy)₃ emission layer in our solution-processed OLEDs because the mCP:Ir(ppy)₃ molecules were completely soluble in chlorobenzene. Toluene and heptane were also investigated as a solvent for the emission layer. However, the mCP:Ir(ppy)₃ molecules were insoluble in heptane and partially soluble in toluene. Since the solvent for the poly-TPD layer was also chlorobenzene, the poly-TPD layer was annealed to increase the resistance to the solvent before spin-coating of the mCP:Ir(ppy)₃ layer. The annealed poly-TPD layer was washed with chlorobenzene solution for investigating the resistance to the solvent. Table 1 shows the thickness and surface rms roughness of the annealed poly-TPD after treatment with chlorobenzene. The annealing was performed at 50, 150, and 220 °C in air. The thickness of the poly-TPD was fixed to be almost 100 nm before chlorobenzene treatment. As expected, the non-annealed poly-TPD film was washed out by the chlorobenzene. Hence, the thickness measured with surface profilometer was ~ 0 nm. The thickness of the poly-TPD film at 50 and 150 °C was

Table 1

Thickness and surface rms roughness after washing with chlorobenzene for the 100 nm poly-TPD layers annealed at various temperatures.

Annealing Temperature (°C)	Thickness (nm)	Surface roughness (nm)
RT	~ 0	4.1
50	~ 0	3.0
150	~ 0	3.3
220	100	0.8

also ~ 0 nm after chlorobenzene treatment, indicating serious dissolution of poly-TPD by chlorobenzene. The non-annealed poly-TPD film exhibits rough surface after washing with chlorobenzene. The surface roughness was 4.1 nm as shown in Table 1. The poly-TPD films annealed at 50 and 150 °C also exhibit rough surface due to the serious dissolution by chlorobenzene. The surface roughness was in the range of 3–4 nm. However, the surface roughness of the poly-TPD film annealed at 220 °C was less than 1 nm. It should be noted that the surface roughness is different for the poly-TPD films without annealing and with annealing at 50, and 150 °C. Although these films were seriously dissolved by the chlorobenzene, we suppose that the very thin partial residues may be on the substrate surface. These partial residues are too thin to detect with the surface profiler. These partial residues may be different from sample to sample. On the other hand, the thickness of the poly-TPD annealed at 220 °C was remained to be almost same after chlorobenzene washing. We also washed the annealed poly-TPD layer with toluene. The poly-TPD film at 220 °C was not dissolved by the toluene solution, either. Fig. 2 shows the surface topologies of the poly-TPD films annealed at 220 °C before and after washing with chlorobenzene. The surface topologies are not seriously changed by the chlorobenzene. The surface roughness of the poly-TPD films annealed at 220 °C was less than 1 nm before and after washing with chlorobenzene. Since the glass transition temperature of poly-TPD is 180–224 °C [17,19,20], the results suggest that the poly-TPD annealed at about glass transition temperature has a high resistance to the chlorobenzene solvent.

Fig. 3 shows the current efficiency as a function of luminance for the solution-processed OLEDs. The device structure was ITO/poly-TPD (45 nm)/mCP:Ir(ppy)₃ (20 nm, 8%)/3TPYMB (50 nm)/LiF/Al. Poly-TPD and mCP:Ir(ppy)₃ layers were deposited by spin-coating. The chlorobenzene was used as a solvent for both the poly-TPD and mCP:Ir(ppy)₃ solutions. The spin-coated poly-TPD layers were annealed at 150 and 220 °C, respectively, before spin-coating of the mCP:Ir(ppy)₃ layer. The maximum current efficiencies were 7.7 and 39.9 cd/A for the devices with poly-TPDs annealed at 150 and 220 °C, respectively. The

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