



The effects of solvent vapor annealing on the performance of blue polymer light-emitting diodes



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ARTICLE INFO

Article history:

Received 28 June 2015
Received in revised form
24 August 2015
Accepted 24 August 2015
Available online xxx

Keywords:

Polymer light-emitting diodes
Solvent vapors annealing
Film morphology

ABSTRACT

In this manuscript, we reported the effects of solvent vapor annealing on performances of blue polymer light-emitting diodes, where the blue light-emitting poly(dibenzothiophene-*S,S*-dioxide-co-dioctyl-2,7-fluorene) that comprising dibenzothiophene-*S,S*-dioxide with molar ratio of 10% was used as the emissive layer, and toluene or chloroform was use as the annealing solvents. It was noted that the performances of PLED in terms of both luminous efficiency and brightness were obviously enhanced by solvent vapor annealing treatment, which can be correlated to the improved film morphology. The formation of β -phase lead to more balanced electron and hole flux as evaluated by single carrier devices. The best device performance with luminous efficiency of 4.8 cd/A was realized, which increased about 70% relative to that of 2.77 cd/A for the pristine device. These observations demonstrated that the solvent vapor annealing approach can be an effective strategy to improve performances of polymer light-emitting diodes.

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1. Introduction

Since the first report of polymer light-emitting diodes (PLEDs) in 1990, much attention has been attracted by this technology owing to its great promise of fabricating large area, flexible electronic devices based on the low-cost solution processing techniques [1–7]. Despite solution-based processing conditions of PLEDs have specific advantages in comparison to the vacuum deposition process [8–13], the electroluminescent performances of PLEDs are lagged behind the small molecules based organic light-emitting diodes (OLEDs) [14–17]. Therefore, developing highly efficient solution processable PLEDs is of particular importance for the community.

Recent progress in light-emitting polymers presents a range of novel species that can lead to efficient light-emission along with excellent spectra stabilities. With respect to the green- and red-light-emitting polymers, however, highly efficient and stable blue light-emitting polymers remain a great challenge. It has been

reported that by introducing the electron-deficient unit, such as dibenzothiophene-*S,S*-dioxide (SO), into the backbone of polyfluorene derivatives can lead to highly efficient blue-light-emitting conjugated polymers with outstanding spectral stability and color purity [18–26].

In addition to the access of highly efficient blue light-emitting conjugated polymers by chemically modified molecular architectures, the alternative strategies, such as the optimization of device architectures or film morphology to attain balanced charge carrier transportation that can in turn result in excellent performances, are attracting increasing interests. Although it has been well-established that the thermal annealing of the emissive layer can be an effective approach to improve the performance of polymer light-emitting diodes [27–31], the polymer films essentially take a risk of potential degradation at relatively high thermal annealing temperatures and thereby detrimental to the device performances [32–38].

In this respect, the solvent vapor annealing technique, which is carried out at room temperature, exhibits great potential to attain high performance by improving intermolecular stacking of polymer main chain while retaining the film morphology stability [39–44]. On the basis of this approach, Liu et al. have treated the MEH-PPV film by using the solvent vapor of tetrahydrofuran and toluene, and achieved more than 10% improvement in luminous efficiency with respect to the pristine device [45]. Measurements of the solvent

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vapor treated polymer films illustrated that the film thickness varies in roughly linear fashion with pressure, while in inverse fashion with temperature, demonstrating that the fundamental reason can be correlated to the solvent-assisted dye diffusion in the emissive layer [36]. It was also noted that the thickness can increase up to 15% with respect to the pristine film after solvent annealing [46,47]. In addition to the improved film morphology, the more balanced charge injection can also be attained by solvent vapor annealing due to the improved interface between the polymer and cathode [48].

In this work, we systematically investigated the effects of solvent vapor annealing approach on the electroluminescent performances of PLEDs based on the blue light-emitting polymer poly[(dibenzothiothiophene-*S,S*-dioxide)-*co*-(dioctyl-2,7-fluorene)] (PFSO10) as the emissive layer, where chloroform and toluene were utilized as the annealing solvents due to their good dissolubility for PFSO10. It was found that the electroluminescence spectra were nearly unchanged while the light-emitting efficiency can be obviously enhanced after solvent annealing with respect to the pristine films, which can be primarily correlated to the formation of β -phase and improved film morphology along with more balanced hole/electron flux in the emissive layer. The best device performance with the maximum luminous efficiency of 4.84 cd/A was achieved with the International Commission on Illumination (CIE) coordinates of (0.16, 0.14).

2. Experiments

The poly[(dibenzothiothiophene-*S,S*-dioxide)-*co*-(dioctyl-2,7-fluorene)] (PFSO10) used in this study comprised 10 mol% of dibenzothiothiophene-*S,S*-dioxide, which was synthesized according to the previous reported procedure (molecular structure shown in Fig. 1a) [49]. Solvent vapor annealing process was carried out at room temperature in a Petri dish filled with solvent vapors. The employed annealing solvents are chloroform and toluene, both of which are good solvents for PFSO10. In order to produce a solvent vapors atmosphere, the solvent (2 mL) was initially added into a pre-cleaned Petri dish for 5 min. Then the fabricated polymer films were transferred into the Petri dish for a certain amount of time. After the solvent vapor annealing, all polymer films were pumped for 2 h under 10^{-1} Pa to remove the residual solvent before the deposition of cathode. Detailed process of solvent vapor annealing is shown in Fig. 1 (b).

The fabrication of the devices followed a well-established

process. The ITO glass substrates (sheet resistance of 15–20 Ω /square) were successively cleaned in an ultrasonic bath by using acetone, detergent, deionized water, and isopropanol. The ITO glass substrates were dried in vacuum oven overnight at 80 °C. A PEDOT:PSS layer (40 nm) was spin-coated directly on the ITO substrate after oxygen plasma treatment. The PEDOT:PSS was heat treated at 120 °C for 20 min. The emissive layer of PFSO10 was spin-coated from *p*-xylene solution and then annealed at 100 °C on a hotplate for 20 min. The thickness of ~80 nm was determined by a Tencor Alpha-step 500 Surface Profilometer. For the cathode, 1.5 nm of CsF followed by 120 nm of aluminum (thickness monitored with a STM-100/MF Sycon quartz crystal) were deposited by thermally evaporating at a base pressure of 1×10^{-6} Pa. The pixel area (16 mm²) was defined by a shadow mask between the cathode and anode. Except for the deposition of PEDOT:PSS layer, all other procedures were carried out in the nitrogen filled glove-box with the oxygen and moisture below 10 ppm. The PLEDs were encapsulated with a UV-cured epoxy resin.

UV–vis absorption of the films were recorded on a HP 8453 spectrophotometer and photoluminescence spectra on a spectrofluorimeter. The morphology of the polymer film was characterized using atomic force microscopy (AFM). The images was recorded in tapping mode on a Seiko SPA 400 equipped with an SPI 3800 probe station. The luminance – current density – luminous efficiency ($L - J - LE$) characteristics were collected by using a Keithley 236 source measurement unit and a calibrated silicon photodiode. The luminance was calibrated by a PR-705 Spectra Scan spectrophotometer (Photo Research), with simultaneous acquisition of the EL spectra and CIE coordinate, driven by Keithley model 2400 voltage–current source.

3. Results and discussion

Fig. 1 shows the chemical structure of poly[(dibenzothiothiophene-*S,S*-dioxide)-*co*-(dioctyl-2,7-fluorene)] (PFSO10) (see in Fig. 1) a and the schematic illustration of solvent vapor annealing procedure used in this study (see in Fig. 1b). The blue light-emitting PFSO10 comprises 10 mol% of dibenzothiothiophene-*S,S*-dioxide (SO) moiety. The emissive layer of PFSO10 was fabricated by spin-coating from the *p*-xylenes solution, followed by treating in a Petri dish filled with solvent vapors. The UV–vis absorption and photoluminescence (PL) spectra of PFSO10 films on the top of quartz are shown in Fig. 2. The maximum absorption of PFSO10 films before and after solvent vapor annealing are located at about 385 nm. In addition, a small shoulder at about 430 nm was observed for the solvent vapor annealed films, which was most likely due to the formation of β -phase from PFSO10 film after solvent vapor annealing [50]. The PL spectra of all films exhibited two peaks located at 440 nm (0–0 band) and 467 nm (0–1 band), and the peak located at 467 nm become stronger after solvent vapor annealing, which can be attributed to more effective energy transfer from amorphous state to β -phase due to polymer chain rearrangement [51]. It is also worth noting that there is a slight differences in the shoulder emission of Fig. 2(a) and (c) at about 430 nm, where the peaks for toluene based samples are not as sharp as that of chloroform annealed counterparts. However, since the lack of high temperature annealing procedure, it is not sufficient to attribute these signals to the semicrystalline state of poly(9,9-dioctylfluorene)s. Instead, we suppose that such differences can be ascribed to the different ratios of β -phase in the solvent vapor annealed films. To verify this assumption, we carried out the peak deconvolution of the absorbance after solvent vapor annealing. The estimated content of the β -phase is about 7.0% for chloroform vapor annealing films, which is higher than that of about 5.7% for toluene vapor annealing films. These observations

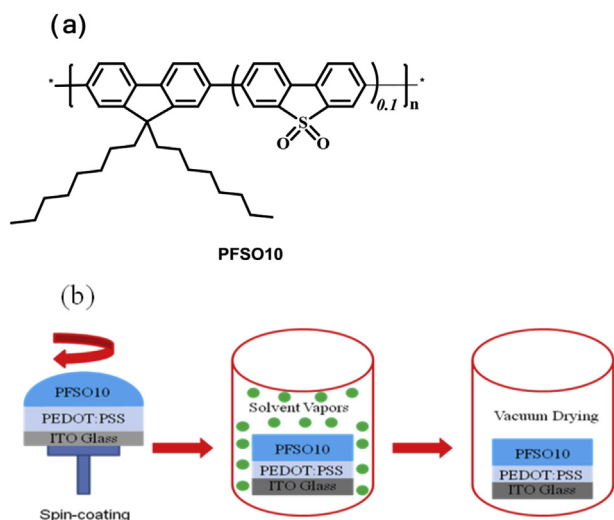


Fig. 1. Chemical structure of PFSO10 (a) and the illustration of normal spin-coating and solvent vapor annealing process (b).

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