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Luminescent Spectral Changes in Polymer Light-Emitting Diodes after Heat Treatments

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Luminescent spectra of polymer light-emitting diodes (LEDs) would depend upon the heat treatment condition and procedure greatly. We have tried thermal annealing at the temperature above T_g of poly[2-methoxy-5-(2′-ethyl-hexyloxy)-1,4-phenylene vinylene] (MEH-PPV), before and after Al deposition respectively, in ITO/MEH-PPV/Al structure. The pre-deposition annealing makes the PL and EL spectrum red-shifted and more broadened. After annealing, the optical band-gap of MEH-PPV thin film decreases by about 0.1 eV relative to that of the dilute solution. The annealing also makes an excimer emission dominant in PL and EL spectra. In contrast, after post-deposition annealing, the EL spectrum was blue-shifted and narrowed. In addition, the excimer emission is withdrawn remarkably after the post-annealing.

Keywords: Electroluminescence; Spectral change; Heat treatment

INTRODUCTION

There has been considerable progress in polymer electroluminescent (EL) devices since the electroluminescence from conjugated polymers

discovered [1]. The optical and luminescent properties usually depend on the molecular arrangement [2]. In addition, inter-chain interaction in conjugated polymers makes a critical impact on the luminescent properties of ordered thin film [2]. On the other hand, the EL output and efficiency are greatly dependent on the heat treatment procedures and temperature [3].

In this article, we investigated the heat treatment effect on the luminescent spectra by treating thermal annealing (heat treatments above T_g) on the thin film for EL devices.

RESULTS AND DISCUSSION

The optical absorption of poly[2-methoxy-5-(2'-ethyl-hexyloxy)-1,4-phenylene vinylene] (MEH-PPV) (T_g=83°C) in dilute solution and thin film is shown in Figure 1. In solution, the onset of optical absorption is 2.1 eV. The onset point of optical absorption in thin solid film changed to be lower energy and the FWHM (full width at half maximum) of the absorption spectrum is more broadened. In annealed films above T_g, more spectral broadening is observed compared with the unannealed film and the optical badgap is narrower by ~0.1 eV relative to that of the solution. We attribute this narrowed bandgap to the increase of the effective conjugation length and the broadened FWHM the redistribution of conjugation length by the chain rearrangement within the emissive polymer. It is well known that cofacial chain packing in thin film influences its optical and luminescent property [2]. The annealed films are more closely packed than unannealed film and show different optical and luminescent properties. The chain packing was

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confirmed by the reduction of thickness after the thermal annealing.

Figure 1 also shows the photoluminescence (PL) spectrum of MEH-PPV solution and thin films. In solid films, spectral broadening and red shift in spectrum are observed obviously. The main peak positions of PL spectrum of the solution and the unannealed film are well coincident with the onset of optical absorption (0-0 transition), while those of annealed samples are not. The shoulder peak at 625 nm in the unannealed thin film originated from the inter-chain excitation because the peak is not observed in the solution PL spectrum. The shoulder peak grows to be main peak as the annealing temperature increases as shown in Figure 2. The reason for this is that thermal annealing above T_g favors the cofacial chain packing in the solid film.

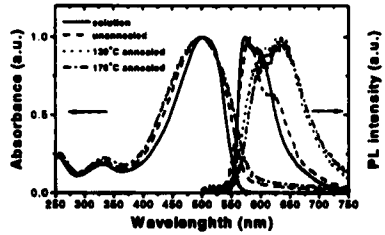

FIGURE 1 The normalized optical absorption and PL intensity of MEH-PPV dilute solution and thin films.

Figure 2 shows the EL spectra in annealed devices before and after Al deposition. Thermal annealing greatly changes the EL spectra as well as the PL spectra. The EL spectrum of annealed devices before Al deposition is similar to the PL spectrum in the point of the red-shift and FWHM broadening. The EL spectrum after the pre-deposition annealing also is also greatly influenced by the physical dimer or excimer, which results from inter-chain interaction. The PL and EL

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efficiency tends to be lower after pre-deposition annealing. Therefore, thermal annealing before Al deposition must be carefully considered. However, the EL maximum output increases due to the enhanced thermal endurance after annealing. Thermal annealing after Al deposition makes the EL spectrum blue-shifted and narrowed in contrast to the red-shift and broadening in spectrum before Al deposition. In addition, the EL spectrum does not show any excimer contribution after 170°C annealing. The EL maximum output of post-deposition annealed device at 170°C is about 10 times greater than the unannealed one [3]. Therefore, it can be said that rather intrinsic intra-chain excitation than inter-chain occurs dominantly after post-deposition annealing, which results in a narrower spectrum with the EL maximum similar to the solution PL maximum. As a result, the post-deposition annealing is recommended as a thermal treatment for an bright and efficient EL device.

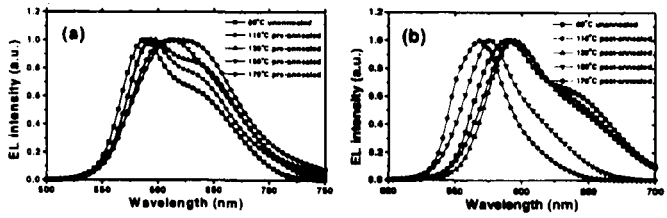

FIGURE 2 The normalized EL spectrum (a) before and (b) after Al deposition.

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