

High efficiency polymeric light-emitting diodes with a blocking layer

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Abstract

In this paper, we describe a simple method for fabrication of high efficiency multilayered polymeric light-emitting diodes (PLEDs). In the device configuration of ITO/HIL/BL/EML/BaF₂:Ca:Al (ITO: indium tin oxide, BL: blocking layer, HIL: hole injection layer as PEDOT:PSS, EML: emitting layer as poly(alkylfluorene) derivative), poly(9-vinyl carbazole) (PVK) was selected as an BL because it has a hole transporting property and a relatively higher band gap, especially a lower LUMO level than the emitting polymer and higher HOMO level than PEDOT:PSS. As a result, the PVK layer could effectively block the electrons from metal cathode and confine them in the emitting polymer layer. In addition, it could properly control the amount of holes from ITO anode by partial blocking. The charge balance was achieved and the device with a PVK layer exhibits a higher brightness and luminance efficiency than those in devices without PVK. Also, the thickness of PVK was easily controlled by solvents for emitting polymer and the device with a thicker PVK layer shows better performance.

Keywords: organic light-emitting device, conjugated polymer, charge balance, blocking layer

1. Introduction

Organic light-emitting devices (OLEDs) have received a lot of attention for their application as flat panel and full colour displays [1,2]. Since the basic structure of OLEDs which is composed of thin film of sandwiched organic emitting materials between two electrodes has been introduced, more multiple functional organic layer structured devices, which provide separated functions such as hole transporting, injection, electron transporting and injection, have been developed to further improve the performance of devices. It seems that for OLEDs, the more layers there are in the device structure, the better the device performance. In the small molecular device, this multilayer can be easily fabricated by multiple vapor deposition methods. However, the multilayer fabrication in the polymeric device is not easy, even layers were fabricated by simple film preparation methods such as a spin coating or ink-jet, because the first layer needs to be solvent resistant to enable the fabrication of the subsequent layer. As a result, just one additional layer, which is a poly(3,4-ethylene dioxathiophene): poly(styrene sulfonic acid) (PEDOT:PSS), as a hole injection layer can be commonly used in the current polymeric LEDs (PLEDs).

It was well-known that the efficiency of OLEDs depends on the carrier injection and carrier recombination efficiency. The balance of holes and electrons is required for obtaining high carrier recombination efficiency and thus high device performance [3,4]. A number of attempts have been made to achieve charge balance of holes and electrons [5,6,7]. Among them, an inserting of thin layer of organic conducting, semiconducting or insulating materials such as polyaniline, [6] poly(phenylenevinylene), [6] and teflon, [7] between the electrode and the emitting layer was used as simple methods to increase the device efficiency. In these studies, it seems that the excess holes from anode were controlled (blocked) by inserted thin layers which may help improve the balance of hole and electron injections.

Another approach improves device performances by blocking electrons has been more attracted recently. [8] D. Heithecker *et al.* reported the electron blocking at the Alq₃/hole transporting interface and therefore the carrier are confined in the emitting zone improving EL characteristics [8].

In this report, we have developed multilayered PLEDs which have an additional layer between the hole transporting and emitting layers. This layer used to not only control the amount of holes and help charge balances but

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also to block the electrons, electron blocking layer (EBL). Poly(9-vinyl carbazole) (PVK) was chosen as the material for the layer because it was known as a good hole transporting and wide bandgap material but not an electron transporting material. By careful selection of solvents and spin processing, PVK and followed emitting layers can be successfully fabricated without severe damages in morphology of PVK layer by emitting polymer solution. This PLEDs show better device performance in a brightness and efficiency.

2. Experimental

A emitting polymer, poly(alkylfluorene) derivative (PF) was synthesised by the method in the literature [9] PVK ($M_n=100000$ g/mole) was obtained from Aldrich and used without further purification. PEDOT:PSS (Baytron-P) was purchased from Bayer. For spectroscopy measurements, thin films were prepared by spin coating from solution with toluene as solvent on quartz substrates. The UV-vis absorption spectra were measured with a HP 8453 spectrophotometer. Photoluminescence (PL) spectra were collected with a Fluorolog 3 double grating spectrofluorometer (Jobin-Yvon Spec Corp.). Cyclic voltammetry (CV) experiments were performed with a BAS-100 electrochemical analyzer. All measurements were carried out a glassy carbon working electrode, a platinum auxiliary electrode and a non-aqueous Ag/AgNO₃ reference electrode.

The device configuration is ITO/PEDOT:PSS(65 nm)/PVK(10–20 nm)/PF(70 nm)/BaF₂(2 nm)/Ca(50 nm)/Al(300 nm). PEDOT:PSS was spin coated onto a pre-cleaned ITO surface and baked at 110 °C for 10 min. The PVK solution was prepared with chlorobenzene and then spin coated onto the surface of PEDOT. This film was baked at 180 °C for 1 hr. In the same way, the emitting polymer was then spun onto PVK layer followed by baking at 160 °C for 1 hr. The several solvents such as toluene, chlorobenzene and trichloroethane were tested for the emitting polymer to find the condition for a good PVK/Emitting polymer layer configuration. The PVK coated glass substrate was rotated on the spin coater and the PVK film slowly washed by solvents. After baking at 160 °C for 1 hr, the thickness and surface morphology of PVK film were measured by surface profiler (TECOR P-10) and atomic force microscopy (AFM, Nanoscope IIIa, Digital Instruments Co.).

The BaF₂/Ca/Al cathode was deposited through a shadow mask by thermal evaporation at 3×10^{-7} torr. Finally, this device was encapsulated by the metal can with dry agents. The device in the similar structure without PVK layer was also fabricated to compare. The current-voltage and brightness-voltage characteristics were measured using a Keithley 238 source measurement unit and PR650 (Photo Research Corp.). The electroluminescence spectra were measured with a single-grating monochromator equipped with a photometric charge-coupled device (CCD) camera as a detector.

3. Results and Discussion

Figure 1 shows the generic molecular structure of the emitting polymer. Ar and Ar' denote the aromatic molecules which were connected with poly(dioctylfluorene) units by Suzuki coupling reaction [9]. The molecular weight were estimated to be about $M_n=100000$ g/mole. The absorption and PL spectra of polymer show the absorption peak at 384 nm and emission peak at 474 nm indicating a slightly greenish blue characteristic as shown in Figure 2.

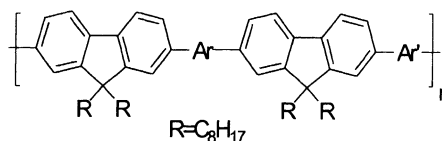


Fig. 1. Generic molecular structure of the emitting polymer.

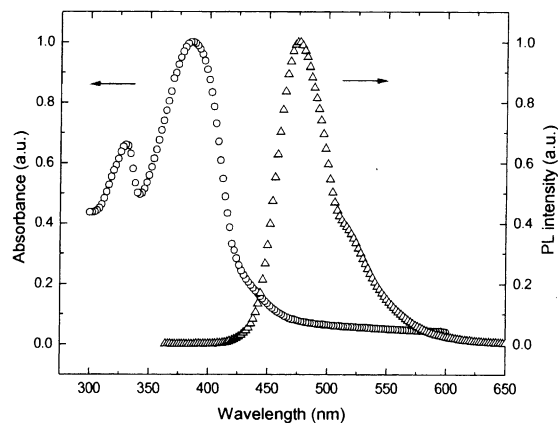


Fig. 2. UV-vis absorption and PL spectra of the emitting polymer.

The electrochemical characteristics of polymer were investigated by the CV method. The first oxidation potential was used to determine the highest occupied molecular orbital (HOMO) energy level. This was then used to obtain the lowest unoccupied molecular orbital (LUMO) energy level with the absorption band edge from UV-vis absorption spectra. Figure 3 schematically depicts the relative HOMO and LUMO energies of the emitting polymer with other layer sequences. Based on these data, PVK can be a good candidate in its function as the electron blocker by the position of its lower LUMO energy level relative to the neighboring PF layer. In addition, the HOMO level of PVK lies beneath that of PEDOT (higher HOMO), thus it can block the hole from ITO electrode as well. This property may help device make the charge balance between holes and electrons.

Since PVK has a role of blocker in both electrons and holes, it is very important to control its thickness. If PVK layer is too thick, the electron can be efficiently blocked and confined in the emitting zone. However, in this case, most of holes may not be injected into emitting polymer

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