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# Molecular Crystals and Liquid Crystals

Publication details, including instructions for authors and subscription information: <a href="http://www.tandfonline.com/loi/gmcl20">http://www.tandfonline.com/loi/gmcl20</a>

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Version of record first published: 18 Oct 2010

To cite this article: Kwang-Jik Lee, Jae-Woong Yu, Jai Kim, Young Kim, Jong Park & Ok Park (2004): Characteristics in Bilayer Organic Light-Emitting Diodes with Variation of the Film Thickness, Molecular Crystals and Liquid Crystals, 424:1, 147-152

To link to this article: <a href="http://dx.doi.org/10.1080/15421400490506081">http://dx.doi.org/10.1080/15421400490506081</a>

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Mol. Cryst. Liq. Cryst., Vol. 424, pp. 147-152, 2004

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# CHARACTERISTICS IN BILAYER ORGANIC LIGHT-EMITTING DIODES WITH VARIATION OF THE FILM THICKNESS

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We fabricated bilayer polymer light-emitting diodes (PLED) with Poly(9,9'-dihexylfluorene-2,7-divinylene-m-phenylenevinylene-stat-p-phenylenevinylene) (PDHFMPPV) and poly(N-vinylcarbazole) (PVK) as an emitting layer and a hole transport layer, respectively. The thickness of each layer was varied, while the total thickness was controlled to be constant as 110 nm. We observed electroluminescence, photoluminescence and current-voltage-luminescence characteristics of these bilayer devices and a single layer device with PDHFMPPV polymer as a light emitting layer for comparison. The quantum efficiencies of the bilayer devices were enhanced up to two about orders magnitude compared with the single-layer device.

Keywords: bilayer; light-emitting diodes; quantum efficiency

Received 1 October 2003; accepted 18 December 2003.

The authors are grateful to the Center for Nanostructured Materials Technology by the  $21\mathrm{C}$  Frontier R&D Program.

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### INTRODUCTION

Organic light emitting diodes (OLEDs) devices become very attractive for their potential applications in large-area, full color, flat panel displays [1]. However, it is highly required to overcome its low efficiency for commercial application. Therefore, several efforts have been carried out, for instance, by changing device structures, inserting additional layers, adding other materials and so on [2,3]. Of these methods, inserting layer plays a role in raising the probability of the electron-hole recombination not only by confining the majority carrier but also by transporting the minority carrier to the emitting layer. It also contributes to the efficiency enhancement by balancing charge injection. Because, for single layer devices, quenching caused by unbalance of the electron-hole injection near the cathode results in low efficiency. Thereby, multi-layer devices are more likely to show the enhanced efficiency than that of the single layer devices [4].

PVK is widely used in PLED as a hole transporting (electron blocking) layer [5,6]. It is also well known for as a donor of Föster-type energy transfer in the blend system [7]. The studies about the best blend ratio of PVK:chromophores were reported elsewhere, while the optimal condition in the layer system is not yet shown [8]. We anticipate that varying the thickness of the polymer and PVK films in bilayer configuration will change the characteristics of the devices.

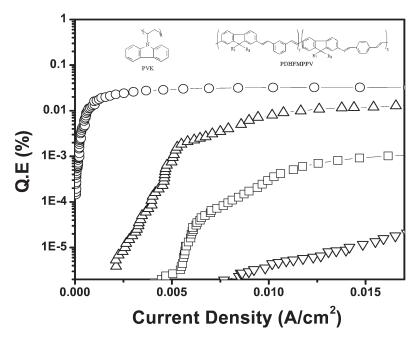
In this work we fabricated bilayer PLEDs with PDHFMPPV and PVK as an emitting layer and hole transporting layer, respectively. The thickness of each layer was varied, while the total thickness was controlled to be constant as 110 nm. The chemical structures of PVK and PDHFMPPV are shown in inset of Figure 1.

Device (a): ITO/PDHFMPPV (110 nm)/Al,

Device (b): ITO/PVK (30 nm)/PDHFMPPV (80 nm)/Al, Device (c): ITO/PVK (60 nm)/PDHFMPPV (50 nm)/Al, Device (d): ITO/PVK (80 nm)/PDHFMPPV (30 nm)/Al.

### **EXPERIMENTAL**

We synthesized the emitting polymeric material, PDHFMPPV, according to the reported synthesis scheme [9]. PVK was purchased from Sigma-Aldrich. PDHFMPPV and PVK were dissolved in Trichloroethylene and Monochlorobenzene, respectively. Film thickness was controlled by the solution concentration and the rate of the spin coater. K.L.A-Tancor:  $\alpha$ -Step 200 was used to measure film thickness. Single layer polymer light emitting device of pure PDHFMPPV and that of the bilayer (PVK/PDHFMPPV) were fabricated as



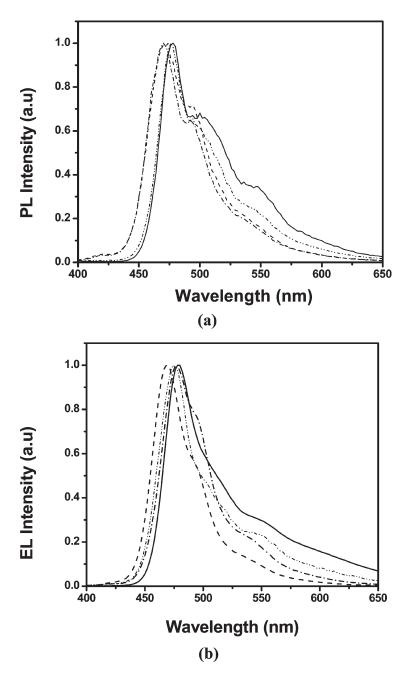
**FIGURE 1** Current density versus quantum-efficiency spectra of ITO/ PDHFMPPV (110 nm)/Al ( $\square$ ), ITO/PVK(30 nm)/PDHFMPPV (80 nm)/Al ( $\nabla$ ), ITO/PVK (60 nm)/PDHFMPPV (50 nm)/Al ( $\triangle$ ) and ITO/PVK(80 nm)/ PDHFMPPV (30 nm)/ Al ( $\bigcirc$ ). The inset shows chemical structures of PVK and PDHFMPPV.

follow: total 110 nm thick layers of PDHFMPPV (110 nm), PVK (30 nm)/PDHFMPPV (80 nm), PVK (60 nm)/PDHFMPPV (50 nm), and PVK (80 nm)/PDHFMPPV (30 nm) were spin cast from each solution on the indium tin oxide (ITO, Samsung Corning) glass. And Al as a cathode was deposited by thermal evaporator under a high vacuum ( $\sim 4 \times 10^{-5}$  Torr).

The current–voltage-luminescence (I-V-L) characteristics of the devices were analyzed using a current/voltage source measurement unit (Keithley 236) and an optical powermeter (Newport 835) connected to a photodiode (Newport 818-SL). Electroluminescence (EL) as well as photoluminescence (PL) spectra were measured by ISS PC1 Photon Counting Spectro-fluorometer.

### RESULTS AND DISCUSSION

Figure 1 shows quantum efficiency (Q.E) versus current density characteristics of the devices. As can be seen, efficiencies of (c) and (d) exhibited much higher values than those of device (a) and (b). Efficiency of the



**FIGURE 2** (a) PL and (b) EL spectra of ITO/PDHFMPPV ( $110\,\mathrm{nm}$ )/Al (solid), ITO/PVK( $80\,\mathrm{nm}$ )/PDHFMPPV ( $30\,\mathrm{nm}$ )/Al (dash), ITO/PVK( $60\,\mathrm{nm}$ )/ PDHFMPPV ( $50\,\mathrm{nm}$ )/Al (dash-dot) and ITO/PVK( $30\,\mathrm{nm}$ )/PDHFMPPV ( $80\,\mathrm{nm}$ )/Al (dash-dot-dot), respectively.

bilayer devices (d) are larger than that of the single layer device up to about two orders magnitude at around  $0.01\,\mathrm{A/cm^2}$ . The enhancement of the quantum efficiency in the bilayer devices can be explained by the following process. One is that energy transfer process from PVK to PDHFMPPV, which will enhance the efficiency. The other is the role of PVK as an electron blocking layer.

Firstly, PVK is well known for as a donor of Föster-type energy transfer [8]. Föster-type energy transfer requires the spectral overlap between the emission of the donor and the absorption of the acceptor. Because a good spectral overlap between the emission of PVK and the absorption of PDHFMPPV was observed Föster-type energy transfer from PVK to PDHFMPPV was expected. To confirm energy transfer, PL intensity of PDHFMPPV neat film was compared with PVK/PDHFMPPV bilayer film. The PVK/PDHFMPPV bilayer film shows much stronger intensity than that of neat polymer. It implies that energy transfer occurs in these devices. Therefore, energy transfer results in enhancements of quantum efficiency.

Secondly, PVK also plays a role in blocking electrons and transporting holes in the device [10,11]. Because PVK provides an electron-blocking effect, most electrons from the cathodes will confined in the PDHFMPPV layer near the interface with PVK. It improves the electron-hole recombination rate of the organic LED. Therefore, quantum efficiency of the bilayer devices is much higher than that of the single layer device. We can notice that quantum efficiency of the devices (b), (c) is lower than of the device (d). This can be explained that PVK thickness of the device (b), (c) is insufficient to offer significant electron-hole recombination.

Figure 2 shows PL and EL spectra of single layer and bilayer devices. As can be seen in figure, there is a little difference between PL, PL EL spectra. The reason for this difference between the PL and EL spectra arises from the different ways that neutral excitations are formed, as described in Ref. [12]. The spectra of bilayer devices showed narrower full width at half maximum (FWHM) compared with that of the single layer device. This behavior can be explained the following: as described above, electron-hole recombination may occur in the PDHFMPPV layer near the interface with PVK. Therefore, we anticipate that PVK molecules and PDHFMPPV molecules can exist inter-mixed state at the interface resulting dilution effect. It can reduce the aggregation of the polymer chains [13]. This introduced much enhanced color purity compared with that of single layer device.

## CONCLUSIONS

Bilayer PLEDs with PVK and PDHFMPPV as hole transporting layer and an emitting layer showed much enhanced color purity and quantum efficiency.

This is attribute to the energy transfer and electron blocking by PVK. Therefore, the thickness effect of the bilayer devices on PLED is considerable to its properties and characteristics.

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