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Thermally Stable Light-Emitting Diodes using the Blend of Polyimide and Poly(9-Vinyl Carbazole) in Hole Transporting Layer

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To obtain a thermally stable organic light-emitting device, the blend film of poly(9-vinylcarbazole) (PVK) and polyimide (PI) having high thermal stability, low thermal expansion, and good adhesion with metal was introduced as the hole-transporting layer (HTL). Overall device structure was glass/anode/ HTL/EML/cathode.

Keywords: organic light-emitting devices (OLEDs); polyimide (PI); hole-transporting layer (HTL); poly(*N*-vinylcarbazole) (PVK)

INTRODUCTION

Since Tang *et al.*[1], who succeeded in fabricating thin film organic light emitting devices (OLEDs) by vapor deposition of functional molecules, inserted the hole transporting layer (HTL) into the device to improve the quantum efficiency, there has been considerable interest in developing OLEDs with high efficiency, good processability, long lifetime and low costs for display applications. Recently, their promising use, large area and flexible display technology came true in some industrial parts. But some difficulties in understanding their mechanism to obtain efficient organic light emitting devices (OLEDs) still remain. In the reported investigations of the fine-tuning balanced charge of OLEDs, the insertion of hole transport layer (HTL) and electron transport layer (ETL) makes the energy band structure balanced. But it is the cause of the Joule's heat responsible for the operational unstability.

In this work, the blend film of poly(9-vinylcarbazole) (PVK) and polyimide (PI) was introduced as a hole-transporting layer (HTL). We discuss the thermal stability of the blend film as an HTL, UV/VIS spectrum, photoluminescence spectrum and current-voltage-brightness (I-V-L) characteristics.

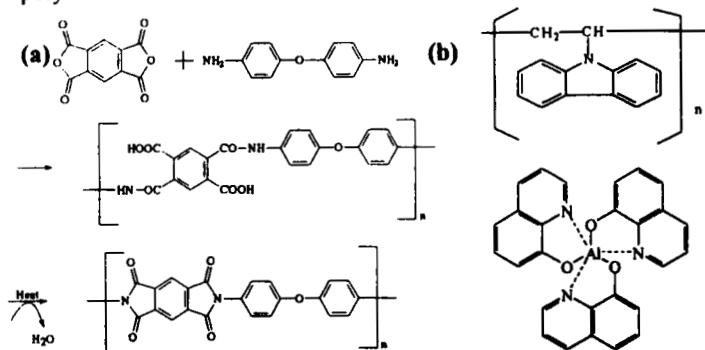
EXPERIMENTAL

We used the blend of poly(9-vinyl carbazole) (PVK) and poly(4,4'-oxydiphenylene pyromellitimide) (PMDA-ODA PI) as an HTL. The device structure of our OLED consists of ITO glass/ blend film/ Alq3/ Al. After spin-casting the blend of PVK and PMDA-ODA PAA onto the ITO glass substrate, it was soft baked at 80°C for 30min and further imidized at 180°C for 1hour under nitrogen atmosphere. Pure PVK film was annealed at 80°C for 30min. Scheme 1 illustrates the thermal imidization process (a) and chemical structures of PVK and Alq3 (b). The emitter, tris-(8-hydroxyquinoline) aluminum (Alq3) was vacuum-deposited onto the blend film of PVK and PMDA-ODA PI. The cathode material (Al) was evaporated on the top of Alq3 layer. All the thermal evaporation process was performed under the vacuum of 2×10^{-5} Torr (the active area: $0.2\text{mm} \times 0.2\text{mm}$). The EL spectra were measured using S2000 fiber optic spectrometer (Ocean Optics Inc.). The photoluminescence spectrum was obtained using fluorescence spectrometer (SFM25, KONTRON Co. Ltd.). The current density-voltage-luminescence (J-V-L) characteristics was measured with a photomultiplier tube (PMT) (Hamamatsu Photonics Co.) and an electrometer (Keithley 6517) controlled with a personal computer via analog-to-digital converter (ADC) and IEEE488 GP-IB card. All measurements were carried out under air ambient and at room temperature condition.

RESULTS AND DISCUSSION

As stated in the introduction section, the thermal stability of all device elements is an important requirement to assure operational stability of OLEDs [2]. In the present system, PVK and PMDA-ODA PI have high glass transition temperatures, about 220°C and 377°C, respectively. It is noteworthy that our OLED was fabricated utilizing thermally stable

polymer blend film of PVK and PMDA-ODA PI as HTL.



SCHEME 1. (a) Polymerization of poly (amic acid) and thermal imidization process and (b) structure of poly(9-vinylcarbazole) and Alq3

After spin-casting process, the induced residual stress presents in the thin film because the thickness of the polymer layer is as nearly same as or smaller than the radius of gyration $\langle S^2 \rangle^{1/2}$ of polymers. Figure 1 shows differential thermal analysis (DTA) data of the polymers. The pure PVK film was quite unstable as compared to the blend film under thermal treatments. It is assumed that the blend film is suitable for enduring operational heat. However, care should be taken to eliminate the mismatch in the residual stress after spin-casting through the thermal treatments such as soft-baking and thermal imidization, in order to guarantee the durability of the OLEDs.

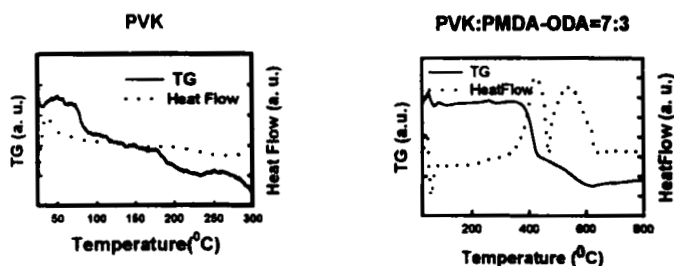


FIGURE 1. Differential thermal analysis (DTA) data of pure PVK thin film and the blend film of PVK and PMDA-ODA under thermal treatments.

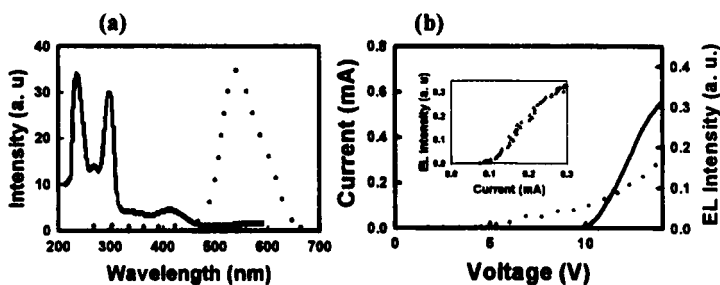


FIGURE 2. UV/VIS absorption spectrum (solid) and photoluminescence spectrum (dotted) of ITO/blend film/Alq3 (a) and current-voltage-EL intensity characteristics (b).

Figure 2 shows the UV/VIS absorption spectra of device elements and characteristics of current density-voltage-EL intensity of the OLED utilizing PVK and PI blend film. The absorption peaks of ITO/blend film of PVK and PI/Alq3 were observed at 400nm, 280nm, and 210nm. The photoluminescent peak was observed at 520nm. In the current and EL intensity relationship, we could find the threshold. It seems that electrons injected through the Al are blocked by HTL during the initial operating. The driving voltage was about 10V and the bright green light emitted from the device at 13V was clearly visible and stable under ordinary room light. Further study is now underway on the stability of devices used in this work including lifetime measurements.

Acknowledgments

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