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## **Stable Low Voltage Organic Light-emitting Diode Using Insulating Polymer/Conducting Polymer Blends**

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Organic light-emitting diodes (OLEDs) utilizing the blend of doped polyaniline (PANI-DBSA) and aromatic polyimide (PMDA-ODA PI) as a hole transport layer (HTL) were successfully fabricated. Tris(8-hydroxyquinolato aluminum) ( $\text{Alq}_3$ ), indium-tin oxide (ITO), and aluminum (Al) were used as a lumophore, transparent anode, and stable cathode, respectively. The charge injection of the OLEDs with the softbaked and imidized HTL was started at *ca.* 4 Vdc and 3 Vdc, whereas the turn-on voltage was *ca.* 5 Vdc for the OLED with the imidized film as a HTL. The electroluminescent (EL) spectra of both OLEDs were almost the same as those of photoluminescence (PL) of the HTL/ $\text{Alq}_3$  films with marginal shift toward higher energy.

**Keywords:** OLED, polyaniline, polyimide, hole transport layer,  $\text{Alq}_3$

## INTRODUCTION

Polyimides have been used for the OLED to enhance the device durability due to their outstanding properties such as high thermal stability, low thermal expansion, good solvent resistance, and high mechanical strength<sup>[1-3]</sup>. In these reports, the polyimide was used as a host polymer binding lumophores or hole transport molecules. However, the polyimide could not contribute to the decrease of turn-on voltage because of its inherent insulating characteristics. Thus it is required to be modified with other conducting materials to have an electrical conductivity above semiconductor level for a stable device with lower energy consumption. Since the doped polyaniline has both of relatively high conductivity and hole injection ability, it has been used as a hole-injecting electrode in OLED<sup>[4-6]</sup>. The first application of conducting polyaniline emeraldine salt and soluble polyimide blend for the OLED has been reported, with the turn-on voltage higher than *ca.* 8 Vdc<sup>[7]</sup>.

In the present work, we used for the first time, to our best knowledge, the blend of doped conducting polyaniline emeraldine salt and insoluble fully aromatic polyimide as a hole transporting as well as conducting layer for OLED. Core originality of this work is that the vacuum sublimed metal complex dye was used as an electron transporting and emissive layer, leading to the pseudo p-n junction in OLED.

## EXPERIMENTAL

The aromatic polyimide precursor, poly(4,4'-oxydiphenylene pyromellitic acid) (PMDA-ODA PAA), was synthesized with 4,4'-oxyphenylenediamine (ODA) and pyromellitic dianhydride (PMDA) in *N*-methyl-2-pyrrolidone

(NMP) as a solvent under nitrogen atmosphere. Polyaniline emeraldine base (PANI-EB) was prepared from aniline in a well known acidic condition. PANI-EB was converted into polyaniline emeraldine salt (PANI-DBSA) through doping with *p*-dodecylbenzenesulfonic acid (DBSA) in NMP. The solution blend of PANI-DBSA and PMDA-ODA PAA in a weight ratio of 20/80 was carried out at a concentration of *ca.* 1 wt.%. This solution was filtered with 5.0  $\mu\text{m}$  Teflon filter and followed by centrifuge to remove the micro-particles. The final solution was spun onto precleaned ITO-glass lithographed as a 2-line electrode type at 5,000 rpm. The spin-coated thin blend films were softbaked at 85  $^{\circ}\text{C}$ , and thermally imidized at 200  $^{\circ}\text{C}$  for 1 hour under vacuum condition. The chemical structures of PANI-EB, DBSA, PANI-DBSA, PMDA-ODA PAA, and PMDA-ODA PI are shown in Figure 1.

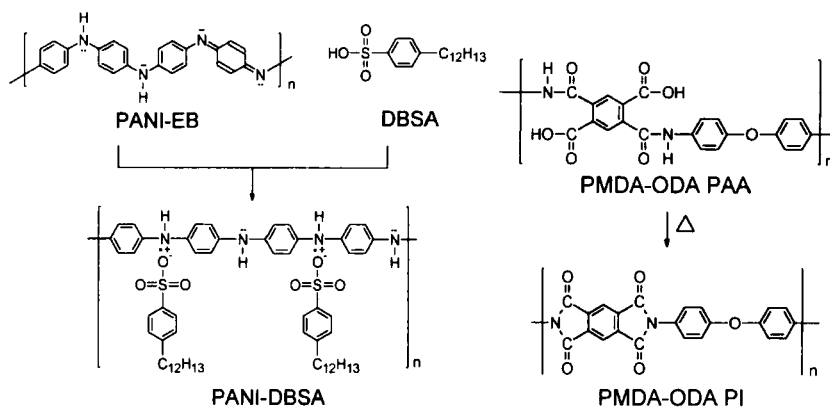


FIGURE 1 Chemical structure of polymers and dopant used.

The softbaked and imidized blend films on the patterned ITO-glass were overcoated with Alq<sub>3</sub> in a vacuum of *ca.*  $4 \times 10^{-6}$  torr at an evaporation rate of

0.02 nm/sec. The thickness of vacuum sublimed  $\text{Alq}_3$  film was *ca.* 5 nm. The Al cathode was deposited onto the  $\text{Alq}_3$  film at a rate of 1 nm/sec until the thickness reached 500 nm. The final device structure was glass/ITO/PANI-DBSA:PMDA-ODA PAA or PI/ $\text{Alq}_3$ /Al. The current density and brightness with applied voltage of the device were measured with Keithley SMU 237 and Si photodiode. The EL and PL spectra were obtained by using JOBIN-YVON 270M Spectrophotometer.

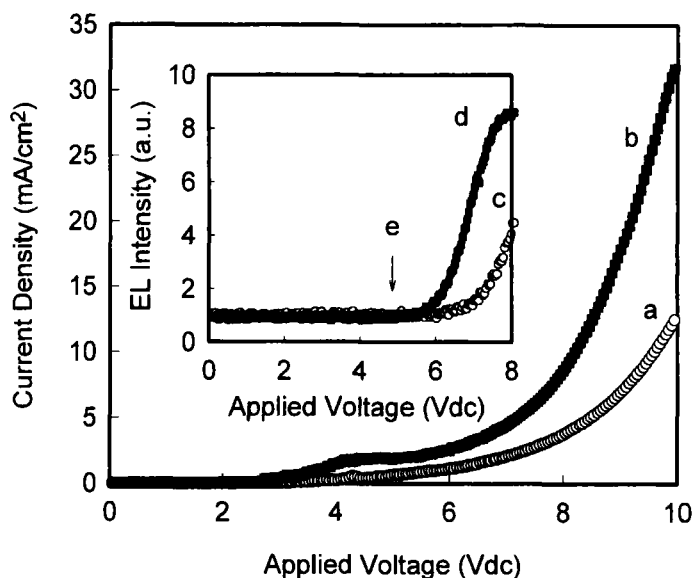


FIGURE 2 Current density and EL intensity characteristics with applied voltage of OLED: softbaked HTL (a,c); imidized HTL (b,d); turn-on point (e).

## RESULTS AND DISCUSSION

The characteristics of current density and EL intensity with applied voltage are shown in Figure 2. It was observed that the charge injection voltages of the OLEDs with softbaked and imidized HTL are *ca.* 4 Vdc and 3 Vdc, respectively. This is due to the decreased resistance caused from the thickness-lowering phenomenon by water molecule extraction *via* thermal imidization. The EL intensity was started to increase at *ca.* 6 Vdc and 5 Vdc for the OLED with softbaked and imidized HTLs, respectively. At these voltages, the green light could be observed with naked eyes. At 8 Vdc, the brightness was above 150 cd/m<sup>2</sup>.

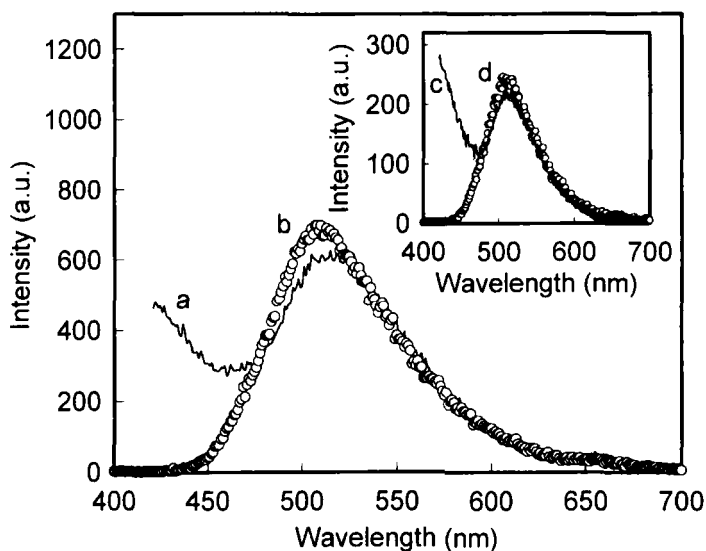


FIGURE 3 PL spectra of softbaked film (c) and imidized film (a) with Alq<sub>3</sub>, and EL spectra of OLED with softbaked HTL (b) and imidized HTL (d).

In Figure 3, the PL spectra, excited at 365 nm, of the softbaked and imidized thin films with the sublimed Alq<sub>3</sub> film are appeared. The EL spectra of the LEDs at 8 Vdc are also shown in Figure 3. The peak wavelength of both of the PL and EL spectra was almost the same at 515 nm (2.4 eV). This energy is originated from the Alq<sub>3</sub> film. Therefore it can be regarded that the softbaked and imidized films do not participate in the emission process but assumed to play a hole injecting or electron blocking role in LED structure.

In conclusion, the OLED using the blend of PANI-DBSA and PMDA-ODA PAA or PI as a HTL was successfully fabricated and emitted green light above *ca.* 5 Vdc. At present, it is believed that the emission mechanism of the OLED is due to the radiative recombination of electrons accumulated in Alq<sub>3</sub> and holes injected from PANI-DBSA/PMDA-ODA PAA or PI layer. The OLED fabricated with the imidized HTL was stable, whereas that with softbaked HTL was not.

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