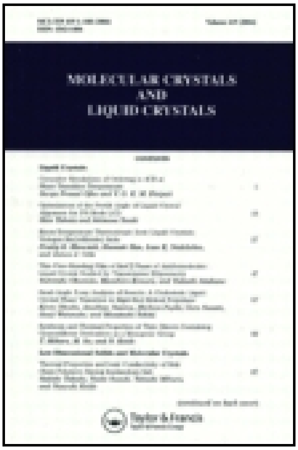

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The Effect of Charge Transport Layers on the Electro-Optic Properties of EL Device with PPV as Emitting Layer

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Abstract : The effect of thermal conversion process of PPV, insertion of poly (N-vinyl carbazole), PVK layer under PPV, and insertion of PBD-polymer matrix as electron transport/hole blocking layer above PPV on the EL device were studied. Luminance intensity of ITO/PVK/PPV/Mg and ITO/PPV/polymer matrix-PBD/Mg EL devices were dependent on the thermal conversion temperature of PPV emitting layer in the order 140℃ > 180℃ > 220℃ > 260℃. Luminance intensity was also dependent on thickness of PVK layer in the ITO/PVK/PPV/Mg EL device. In the ITO/PPV/PBD-polymer matrix/Mg EL device, the intensity of EL device was highest with polystyrene as matrix.

Keyword : Electroluminescence device, poly(p-phenylene vinylene), heterojunction EL device, polymer heterojunction EL device.

INTRODUCTION

Electroluminescence(EL) devices based on organic material are of great interest due to their possible application as large area light-emitting displays. Since Tang et al. succeeded in doubling the electroluminescence (EL) quantum efficiency of molecular organic light emitting diodes[1] by doping tris-(8-hydroxyquinolinate) aluminium (Ⅲ) emitting layer with laser dye, the effects of doping have been the subject of numerous investigations.[2~3] Recent advances in both organic molecular devices and polymeric LED′ s show high potential for EL displays. For molecular devices insertion of heterojunctions between the electrode and the organic electroluminescent layer improved both brightness and operating lifetime.[4] The molecular EL devices have problem of degradation caused by repeated melting and crystallization of organic materials. The polymeric EL device could give solution to these problems and devices prepared with CN-PPV layer showed 4% internal efficiency with Al electrode.[5] This significant improvement could be obtained by the correct balance of ionization potentials which prevent hole/electron recombination at or near the surface of the cathode. The effect of thermal conversion process of PPV, insertion of poly (N-vinyl carbazole), PVK layer under PPV, and insertion of PBD-polymer matrix as electron transport/hole blocking layer above PPV on the EL device.

EXPERIMENTAL

Materials and Synthesis.

α, α' -Dichloro-*p*-xylene and tetrahydro-thiophene were added into methanol and stirred for 19 hr at 50 °C under N_2 atmosphere. The reaction mixture, after cooling, was poured into cold acetone and the precipitate was filtered and dried under vacuum for 48 hr to give *p*-phenylenedimethylene-1,1-bis(tetrahydrothiophene-1-ium)dichloride(yield=66.2%). To the disulfonium salt solution in methanol was added dropwise aqueous NaOH solution over 20 min period at 0°C. The reaction mixture was stirred further for 60 min and a highly viscous PPV precursor solution was obtained. The precursor solution was dialyzed against deionized water at room temperature for 3 days with a cellulose membrane (cut limit ca. 12,000 g/mol). Dialyzed PPV precursor solution was spin coated on the ITO glass. The ITO glass coated with PPV precursor was placed in a vacuum chamber and thermally converted to PPV at different temperature (140 °C ~ 260 °C) for 3 h. Other materials used in this work are shown in Fig. 1.

Fabrication of EL Device.

ITO glass (30Ω/□, 1.08 mm thickness) was cut into 2.5 cm×2.5 cm size and the electrode area was etched by electrolysis method . It was cleaned in a ultrasonic bath with acetone, methanol and mixture of isopropyl alcohol and water (1:1 by vol.). Different types of EL device with PPV as emitting layers were fabricated. First, ITO/PPV/Mg type EL devices in which the thermal conversion temperature of PPV was varied were prepared. Second, for the insertion of PVK layer under PPV emitting layer, PVK solution (different concentration : 0.5, 0.1, 0.05 wt% in dichloroethane) was spin coated on the ITO. It was dried at room temperature for 20 min in the vacuum oven and then PPV precursor aq. solution was spin coated. The PPV layer was thermally converted at different temperature for 3 h.

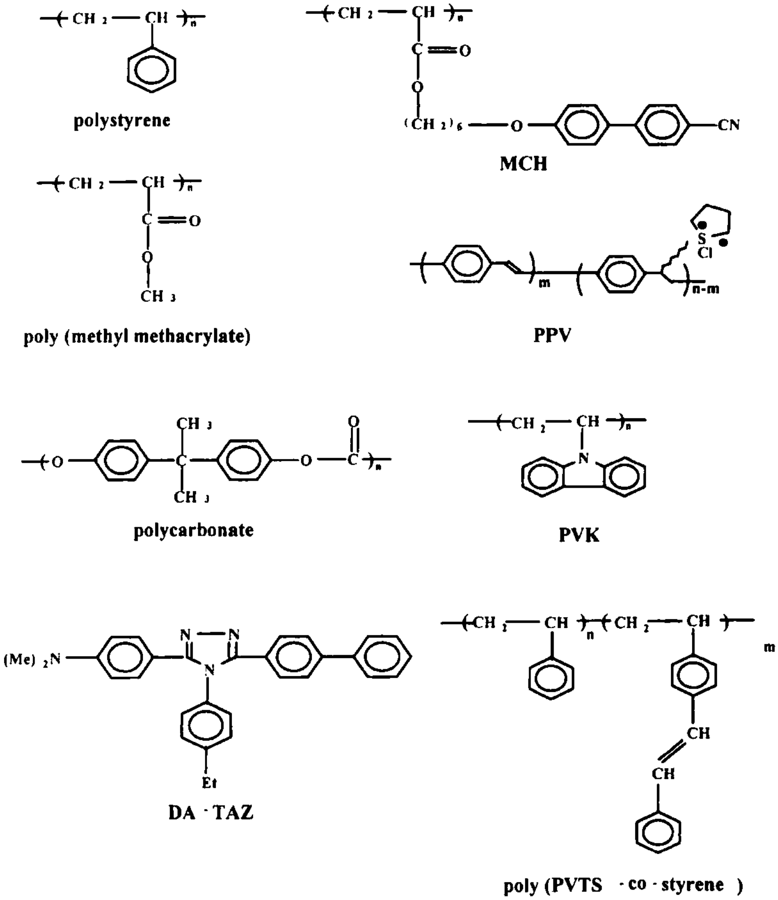

FIGURE 1 Materials used to prepared EL devices.

Third, for insertion of PBD : polymer matrices or DA-TAZ : polymer matrix as electron transport/hole blocking layer, polystyrene, polycarbonate, poly (methyl methacrylate), MCH, and poly (styrene-*co*-PVTS) were used as polymer matrix. PPV precursor aq. solution was spin coated on ITO glass and thermally converted at different temperature (140 °C ~260 °C) for 3h. PBD-polymer matrix layer (different concentration : 0.5, 0.1, 0.05wt% in chloroform) or DA-TAZ : poly (styrene-*co*-PVTS) layer was formed on top of PPV layer by spin coating. The Mg metal electrode was deposited by E-beam method on all of these devices at 12Å /sec rate to 4,000Å thickness.

Characterization and Measurement.

Elemental analyses of monomer and polymers were taken on a Carlo Elba

Model 1106. Current density and luminance were measured with Model 8902A Digital Multimeter and Minolta Luminance Meter LS-100 equipped with close-up lens (No. 110, 40.5mm) at room temperature in the air. Electroluminescence spectrum was obtained by Optical Multichannel Analyzer (EG & G, USA) equipped with OMA4 CCD detector.

RESULT AND DISCUSSION

Electro-optic properties of ITO/PVK/PPV/Mg EL devices.

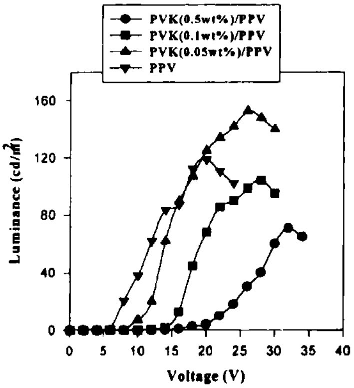

FIGURE 2 Luminance vs. voltage profile of ITO/PVK/PPV/Mg EL devices with different thickness of PVK layer (PPV, 140℃).

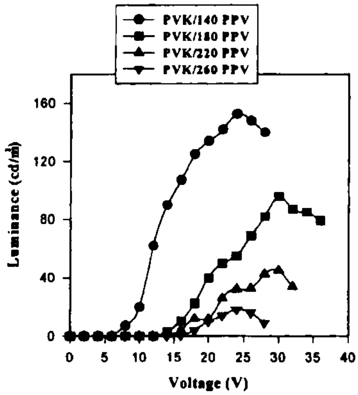

FIGURE 3 Luminance vs. voltage profile of ITO/PVK/PPV/Mg EL devices with PPV layer converted thermally at different temperature.

A thin film of hole transport/electron blocking layer (PVK) was deposited (different concentration ; 0.05, 0.1, 0.5 wt% in dichloroehtane) under PPV layer which was then converted at 140℃. From Fig. 2, these EL devices showed increased threshold voltage compared with ITO/PPV/Mg EL device.

However, the maximum applicable voltage to the device was higher than ITO/PPV/Mg single layer device. EL intensity decreased with the increasing concentration of PVK. This may be due to insufficient hole injection from PVK to PPV layer.[6,7] The maximum luminance of 152.7 cd/m^2 was achieved at 0.05wt% PVK in the ITO/PVK/PPV/Mg devices. ITO/PVK/PPV/Mg hetero-junction EL devices were also fabricated employing different thermal conversion temperature (100 °C ~300 °C) during PPV layer formation. From Fig. 3, it was noted that the threshold voltage of EL device decreased as the thermal conversion temperature to PPV was decreased. The decrease of threshold voltage might be due to presence of some unconverted sulfonium salt in the PPV layer.[8]

ITO/PPV/Polymer matrix : Electron transport agent/Mg EL Devices.

EL devices with electron transport/hole blocking layer on top of PPV emitting layer were fabricated employing electron transport agent (PBD or DA-TAZ) doped in the polymer matrix. The concentration of PBD/polymer matrix in the chloroform solution was varied and the weight ratio of PBD to polymer matrix was 1/1 on the dried film basis. Various polymer such as polystyrene (PS), poly (methyl methacrylate) (PMMA), polycarbonate (PC), and a side chain type liquid crystalline polymer (MCH) were used as matrix polymer in the EL devices. As shown in Fig. 4, EL device with polystyrene as matrix for PBD gave highest luminance intensity of 497.2 cd/m^2 (44.1 mA/cm^2 at 10V). With PS as matrix polymer, the optimum concentration of PBD/PS mixture in chloroform solution was found to be 0.1wt% (Fig. 5).

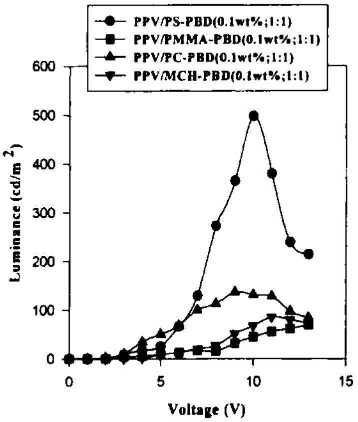

FIGURE 4 Luminance vs. voltage profile of ITO/PPV/polymer: PBD/Mg EL device with different polymer matrix (PPV, 140℃).

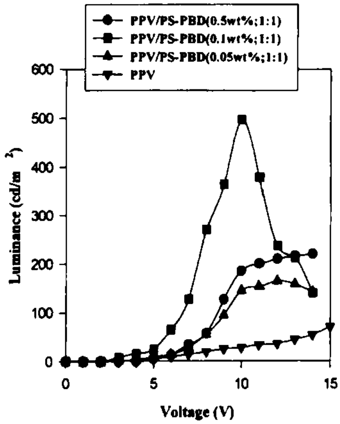

FIGURE 5 Luminance vs. Voltage profile of ITO/PPV/polymer: PBD/Mg EL device with different concentration of PS:PBD (PPV, 140℃).

Since polystyrene as matrix for PBD gave EL device with highest luminance, a copolymer with extended phenyl ring, poly(styrene-co-PVTS), was synthesized. EL devices with poly(styrene-co-PVTS) as matrix material and two electron transport agents (PBD or DA-TAZ) were compared. As shown in Fig. 6 EL device with PBD as electron transport agent gave both higher luminance and stability in the increased electric field.

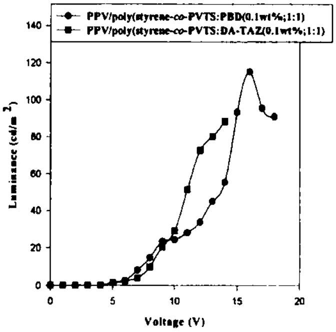

FIGURE 6 Luminance vs. voltage profile EL device with PBD and DA-TAZ as electron transport agent (PPV, 140℃).

CONCLUSIONS

Luminance intensity of ITO/PVK/PPV/Mg and ITO/PPV/polymer matrix-PBD/Mg EL devices was dependent on the thermal conversion temperature of PPV in the order 140℃ > 180℃ > 220℃ > 260℃. Luminance intensity was also dependent on thickness of PVK layer in the ITO/PVK/PPV/Mg EL device. In the ITO/PPV/PBD-polymer matrix/Mg EL device, the intensity of EL device was highest with polystyrene as matrix. Comparison of PBD and DA-TAZ as electron transport agent with poly(styrene-co-PVTS) as matrix polymer showed that PBD was better electron transporting agent than DA-TAZ.

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