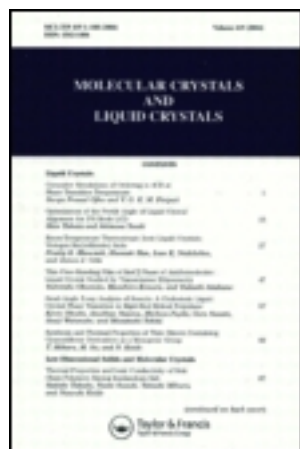


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ELECTROLUMINESCENCE OF DYE-DISPERSED BPDA-PDA POLYIMIDE LIGHT EMITTING DIODE(LED)

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Abstract Photoluminescent(PL) spectra of Alq3 as supplied and after thermal treatment was measured to verify the degradation during thermal imidization of poly(amic acid). The electroluminescent devices(ELDs) composed of ITO, Alq3/BPDA-PDA PAA or BPDA-PDA polyimide, Mg, and In were fabricated. Current-voltage and EL characteristics of the ELDs were also studied. At 50/50 composition, the turn-on voltage of ELD was ca. 12V. The ELD illuminated bright green light at 25V.

INTRODUCTION

Organic light emitting diodes(LEDs) or ELDs have been increasingly studied since vacuum-sublimed dyes and π -conjugated polymers were used as an emission layer. It is difficult, however, to prepare a large area device by vacuum sublimation of the dyes because the size of the vacuum chamber is limited for relatively high vacuum level needed for the ELD fabrication. Even though π -conjugated conducting polymers have been well utilized for ELD, it is not so easy to make thin film onto a substrate because most of them are known to be insoluble in common solvents. Thus, it is of no doubt that the interest moves toward soluble polymers having an electroluminescent chromophores in their main or side chain. Very limited polymers suitable for ELD have been reported except some well-known analogues such as poly(phenylene vinylene)(PPV) derivatives. Nowadays, the dye-dispersed polymers have been used as an emission layer since polymers take an advantage of easy processability, intrinsically good durability and outstanding flexibility in general. For the application of ELD, the host polymers should be thermally stable because the heat generated under high electric field in driving ELD

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owing to ohmic contact between metal and insulator/semiconductor layer could degrade or destroy the device. It was experimentally proved that a polymer having a glass transition temperature below 80°C ¹ is not suitable for ELD. Aromatic polyimide(PI) exhibits high thermal stability, excellent chemical resistivity, and good mechanical properties due to the rigid phenyl and imide moieties of its backbone.² Therefore, it is one of the best candidates as a thermally stable host polymer for ELD. In the present study, the dye-dispersed polyimide ELD were fabricated, and the spectroscopic and EL characteristics were investigated.

EXPERIMENTAL

Figure 1 shows the materials used in this study. Solution blends of Tris(8-hydroxyquinolino)aluminum(Alq3) and poly(*p*-phenylene biphenyltetracarboxamic acid)(BPDA-PDA PAA) in *N*-methyl-2-pyrrolidinone(NMP) were prepared with the composition of 10/90, 30/70, and 50/50 by weight. The solid concentration was ca. 1 wt.%. The solutions were spin-coated at 3000 rpm for 5 min onto a glass for PL measurement and a patterned ITO-glass for ELDs, respectively. The films were softbaked at 80°C for 60 min. The film thickness was 150 - 200 nm. The softbaked films were thermally imidized into poly(*p*-phenylene biphenyltetracarboximide)(BPDA-PDA PI) at 250°C for 60 min as a heating rate of $2\text{K}/\text{min}$. Details of the synthesis of the PI are described elsewhere.² PL spectra of the softbaked and imidized blend films were obtained using fluorescence spectrophotometer(SFM 25, KONTRON Co.Ltd).

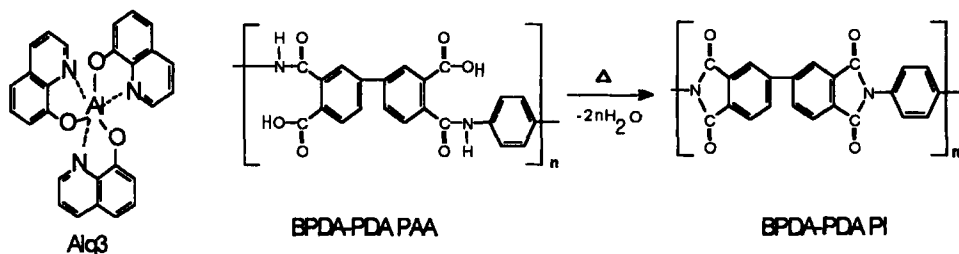


FIGURE 1 Chemical structure of Alq3, BPDA-PDA PAA, and BPDA-PDA PI.

The softbaked and imidized films onto the patterned ITO-glass were overcoated with magnesium(Mg) and indium(In) in a vacuum chamber. The vacuum level and substrate temperature during deposition was below 4×10^{-5} torr and $30 - 35^{\circ}\text{C}$, respectively. The final ELD structures are glass/ITO/Alq3:BPDA-PDA PAA/Mg/In and glass/ITO/Alq3:BPDA-PDA PI/Mg/In.

RESULTS AND DISCUSSION

To verify the thermal stability of Alq3 during thermal imidization of the BPDA-PDA PAA, the PL spectra of Alq3 powders as supplied and after thermal treatment under the thermal imidization condition were measured. The excitation energy was 3.5eV(350nm). As shown in Figure 2-1, no change in the PL shape and the peak position regarded as the degradation is observed before and after thermal treatment. The initial decomposition temperature of Alq3 was measured as ca. 300°C in the thermogravimetric analysis. Thus, it is confirmed that the thermal imidization process doesn't affect to the emission properties of ELDs. Figure 2-2 shows the PL spectra of the BPDA-PDA PAA and the BPDA-PDA PI. The PL of the BPDA-PDA PAA was observed at around 500 nm, while no characteristic PL was detected for the BPDA-PDA PI. At present, no clear spectroscopic explanation can be given for this poly(amic acid) and polyimide. The Alq3/BPDA-PDA PAA(30/70) film shows the slight shift in peak maximum which may be resulted from the constructive effect between Alq3 and BPDA-PDA PAA, not the formation of an exciplex. But the PL peak of the film after thermal imidization is almost the same as that of Alq3 powder.

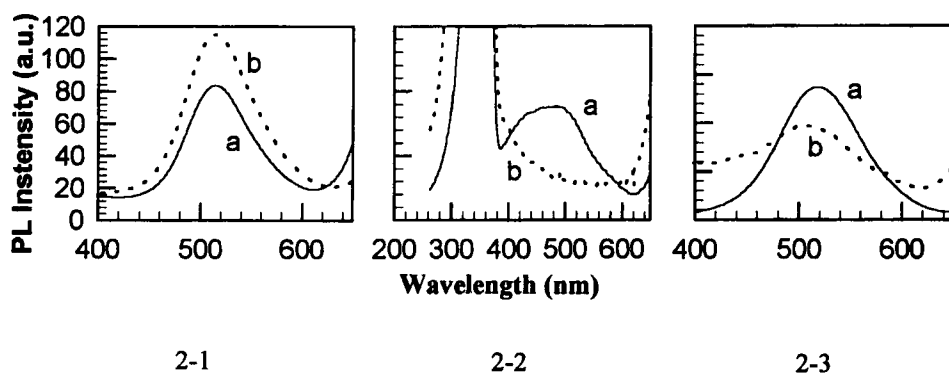


FIGURE 2 (2-1) PL spectra of Alq3 powders as supplied(a) and thermally treated(b), (2-2) BPDA-PDA PAA(a) and BPDA-PDA PI(b), and (2-3) Alq3/BPDA-PDA PAA (30/70)(a) and Alq3/BPDA-PDA PI(30/70)(b) films.

The current-voltage characteristics of the glass/ITO/Alq3:BPDA-PDA PAA/Mg/In ELDs are shown in Figure 3-1. The turn-on voltage of the ELDs is 24V, 18V, and 16V for 10/90, 30/70, and 50/50 composition, respectively. The lower voltage at lower Alq3 content is related to the intrinsically different conductivity of the Alq3/BPDA-PDA PAA films with composition. However, the emitted light was not detected because of very low intensity. It is considered from the current density comparison at the same voltage that the electrons are injected easier in the forward bias than the reverse bias. The deflection of I-V curves between 20V and 40V may be owing to relatively lower thermal stability of poly(amic acid) in the forward bias. In Figure 3-2, the current-voltage relationships of

the glass/ITO/Alq3:BPDA-PDA PAA/Mg/In ELDs are appeared. The current density was increased with Alq3 content. The turn-on voltage at the 50/50 composition was ca. 12V, being lower than that of the ELDs using the BPDA-PDA PAA. It is guessed that the decrement of the turn-on voltage is due to the thinning effect of the aromatic rigid rodlike polyimide film after thermal imidization. Above ca. 15V, the green light was observed. The current density in reverse bias is also less than that in the forward bias. Especially, no light was detected in the reverse bias even at over 20V. This is due to the difficulty in the injection of holes and electrons owing to the potential barrier in the reverse bias. Figure 3-3 shows the PL of the imidized films and EL of the respective ELD at the 50/50 composition of the Alq3/BPDA-PDA PAA. At 18V, the EL spectrum may be vibronic owing to the signal to noise ratio increment. The shape of EL spectrum at 25V resembles the PL, and the wavelength corresponded to peak maximum is almost the same as that of PL. However, the lifetime is ca. 10 min at 25V. This short lifetime is due to the unbalanced injection of holes and electrons, because no hole transport layer was introduced in the present device. In addition to the introduction of the hole transport layer, the effect of the film thickness and the dye composition are also now being undertaken.

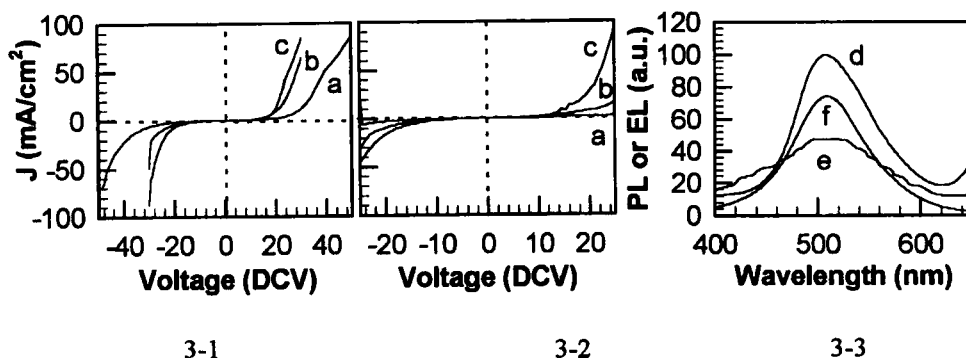


FIGURE 3 Current density(J)-voltage and EL characteristics of ELDs. (3-1) glass/ITO/ Alq3:BPDA-PDA PAA/Mg/In ELD, (3-2) glass/ITO/Alq3:BPDA-PDA PI/Mg/In ELD, (3-3) glass/ITO/Alq3:BPDA-PDA PI/Mg/In ELD. Alq3/BPDA-PDA PAA in 3-1 and 3-2: (a) 10/90, (b) 30/70, (c) 50/50. In 3-3, Alq3/BPDA-PDA PAA (50/50), (d) PL, (e) EL at 18V, (f) EL at 25V.

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