



Molecular Crystals and Liquid Crystals Science and Technology. Section A. Molecular Crystals and Liquid Crystals

Publication details, including instructions for authors and subscription information:

<http://www.tandfonline.com/loi/gmcl19>

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Version of record first published: 04 Oct 2006

To cite this article: Hyuntaek Lim, Hyunjoo Park, Jae-Gyoung Lee, Youngkyoo Kim, Won-jeicho & Chang-Sik Ha (1998): Polyimide/Polyimide Double Layer Structured Light-Emitting Diodes and their Characteristics, Molecular Crystals and Liquid Crystals Science and Technology. Section A. Molecular Crystals and Liquid Crystals, 316:1, 301-304

To link to this article: <http://dx.doi.org/10.1080/10587259808044514>

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Polyimide/Polyimide Double Layer Structured Light-emitting Diodes and Their Characteristics

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Polymeric light-emitting diode (PLED) using polyimide / polyimide double layer structure was prepared in this work. The polyimide was used as a matrix polymer binding a hole-transporting material (HTM) or lumophore. The HTM and lumophore are *N,N'*-diphenyl-*N,N'*-di(m-tolyl)benzidine (TPD) and tris(8-hydroxyquinolinato) aluminum (Alq₃), respectively. The bright green light was observed at 16 Vdc with naked eyes, with the turn-on voltage of *ca.* 10 Vdc.

Keywords: polymeric light-emitting diode (PLED), polyimide / polyimide, poly(ether imide) (PEI), lumophore, hole-transporting material (HTM)

INTRODUCTION

Thermal stability is one of the most important requirements in organic electroluminescent light-emitting diodes (LEDs), because the heat generated during bias makes a severe relaxation of organic or polymeric materials with relatively lower thermal stability than inorganics or metals.

Since aromatic polyimides possess outstanding thermal stability, low thermal

expansion, and high mechanical properties, they have been applied as one of the best candidates to overcome the weakness of organic materials for the polymeric light-emitting diodes (PLEDs)^[1-5]. However, in the previous reports, the emission or hole-transporting layer (HTL) nearby anode was only modified with the polyimide^[1,2,4,5]. Thus it is required that the emitting layer is modified to make the device more stable.

In the present work, both of the HTL and emitting layer (EML) were modified with the high performance polyimide. That is, the homogeneous interface between the same polyimides containing *p*-type and *n*-type organic molecules was made.

EXPERIMENTAL

The PLED structure is anode / HTM-dispersed polyimide / lumophore-dispersed polyimide / cathode. The cross-sectional view of the PLED and the molecular structures of the organic materials used in this study are shown in Figure 1. TPD and Alq3 were used as a HTM and lumophore with electron-transporting ability, respectively. Poly(bisphenol A-*co*-4-nitrophthalic anhydride-*co*-1,3-phenylene diamine) (PEI) was used as a matrix polymer binding the HTM and lumophore.

The solution blend of TPD and PEI in chloroform was prepared by fifty-fifty weight ratio, and the overall solid concentration was 1 wt.%. Alq3 and PEI were also mixed into clear solution with the composition of 70/30 by weight percent in chloroform. The solution of TPD and PEI was spin-coated onto an ITO-coated glass followed by softbaked at 45~50 °C. The mixture of Alq3 and PEI was also spin-coated onto the softbaked TPD-dispersed PEI layer followed by softbaked at the same temperature. Finally, the aluminum electrode was deposited onto the Alq3-dispersed PEI layer in a vacuum of *ca.* 2×10^{-5} Torr. Then the cathode side was packaged with a polymeric glue of poly(ethylene-*co*-vinyl acetate) (EVA) resin at 110 °C. All the measurement was performed

under air ambient condition.

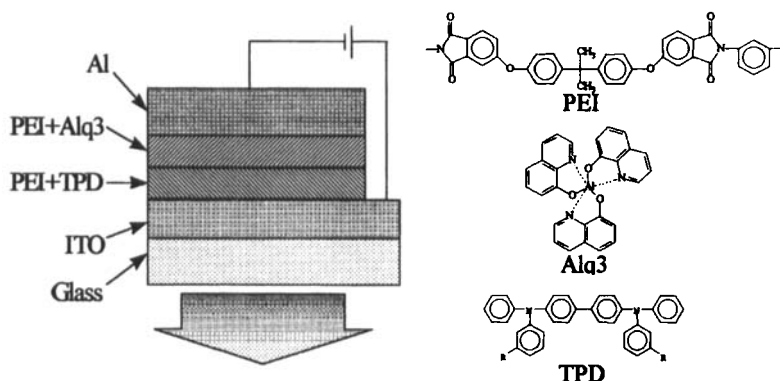


FIGURE 1 The device configuration and the molecular structures of the organic materials

RESULTS AND DISCUSSION

The characteristic curve of current density with bias voltage of the ITO / PEI : TPD / PEI : Alq3 / Al device is shown in Figure 2 (a). The shape of current injection shows typical diode characteristics, meaning the rectified recombination of holes and electrons injected from anode and cathode, respectively. The initial voltage of current injection was *ca.* 10 Vdc. The dependence of EL intensity on bias voltage is appeared in the inset of Figure 2 (a). The turn-on voltage of the device was *ca.* 10~11 Vdc, corresponded to the voltage for the initialization of current injection. It is regarded from the same voltage between current injection and turn-on of light that most of the holes and electrons injected into polyimide-modified *p*-type and *n*-type layer participates in the radiative recombination. In addition, it is also believed that the polyimide/polyimide double layer by spin-coating was successfully prepared with discrete interface between the layers, because the current injection and electroluminescing behaviors may be not accorded and finally go to device

failure.

In Figure 2 (b), the relationship between EL intensity and current density is appeared. It is seen that the EL intensity is linearly increased with the current density. The solid line shows the linear least-squared fitting between the two properties. The relationship coefficient was ca. 0.98, leading to the best fit. From this result, it can be considered that the number of the electrons and holes to be used in the radiative recombination process is almost linearly increased with the applied voltage.

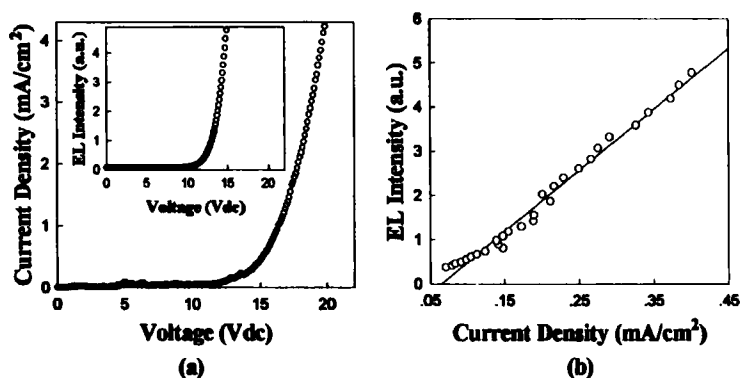


FIGURE 2 Dependence of current density and EL intensity on bias voltage (a); relationship between EL intensity and current density (b) of PLED.

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