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Influence of Thickness and Surface Pressure of Langmuir-Blodgett Films on the Efficiency of Organic Electroluminescence

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The electroluminescent (EL) devices with a single layer structure (ITO/emitting layer/Al) were constructed using LB films of an amphiphilic complex LaL_2Cl for the emitter material. Greenish-yellow EL emission with a luminance of 330 cd/m^2 was observed with a low voltage drive (8 V). The number of the layers of LB films and the deposited surface pressure have strong influence on the I-V characteristics, EL intensity, as well as break-down voltage of the EL devices.

Keywords: electroluminescent devices; Langmuir-Blodgett films

INTRODUCTION

Organic electroluminescent (EL) devices based on organic fluorescent materials are one of the most promising next-generation flat panel display systems. This is because of their potentiality to produce emissions of all colors ranging from blue to red due to the wide selection of organic emitting materials,^[1] which are difficult to make using inorganic light-emitting diodes. In the last decade, the studies of electroluminescence in organic materials have concentrated on the thin-film-type devices made of vacuum-deposited films,^[1,2] or polymer films.^[3] Especially, many complexes of 8-hydroxyquinoline and its derivatives such as Alq_3 , Znq_2 , Beq_2 , Mgq_2 , $\text{Zn}(\text{mq})_2$, and $\text{Al}(\text{prq})_3$, have been used as emitters.^[1,2]

Langmuir-Blodgett (LB) films technique make it possible to prepare organic functional ultrathin films with a controlled thickness at a molecular size and well-defined molecular orientation. Therefore, if amphiphilic complexes with 8-hydroxyquinoline can be incorporated in LB films, the films will be fabricated more conveniently than the general vacuum deposited technique and

may have potential applications in molecular electronic devices. With this in mind, we have synthesized an amphiphilic lanthanum complex with 8-hydroxyquinoline.^[4] In this paper, the I-V characteristics and EL intensity of the organic diode using the LB films of this lanthanum complex as emitter will be studied.

EXPERIMENTAL

The synthesis of the amphiphilic complex, bis[N-hexadecyl-8-hydroxy-2-quinoline carboxamide] lanthanum (LaL_2Cl), and fabrication of LB films were carried out following the previous paper.^[4] The EL device had a single layer structure: ITO/emitting layer (LB films)/Al. The emitting area for diodes was approximately $2 \times 2 \text{ mm}^2$.

RESULTS AND DISCUSSION

The emission from the LaL_2Cl LB films was observed for the organic EL diodes with continuous DC for a forward-bias ITO as positive polarity. Fig. 1 shows the relationship of current density (I) and voltage (V) in the EL devices. The shapes of the I-V curves are strongly dependent on the thickness of the LB films of LaL_2Cl (luminescence layer). With increasing LB film layers, the driving voltage increases. The driving voltages for the devices with an emitter of 9-, 19- and 29-layer LB films are ca. 8, 9 and 10 V, respectively. The I-V curves can be fitted to an injection-limited model where the electron current is limited by electron injection from the cathode into the LaL_2Cl LB film layer.^[5]

Fig. 2 shows the luminance-current density characteristics of the EL device with an emitter of 9-, 19- and 29-layer LB films. The luminance is proportional to the injection current in the region of $0.2\text{--}0.8 \text{ mA/mm}^2$. The thickness of luminescent layer (LB films) drastically influences the emission of the EL devices. The luminance for the device with 19-layer LB films (47.5 nm) is larger than those with 9-(22.5 nm) and 29-layer (72.5 nm) LB films. It can be interpreted that the luminance (B) is directly proportional to the carrier (electron and hole) concentration (n_e and n_h) and the electron-hole radiative recombination probability (γ).^[3]

$$B \propto \gamma \cdot n_e \cdot n_h \quad (1)$$

When LaL_2Cl LB films were used as an emitter, the recombination zone was located within about 6-layers of the LB films (ca. 10 nm) adjacent to the ITO to a distance of about 9-layer LB films (ca. 22.5 nm).^[6] In the EL device with 9-

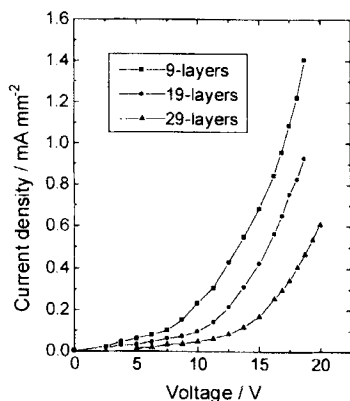


FIGURE 1 I-V characteristics for different layer LaL_2Cl LB films devices.

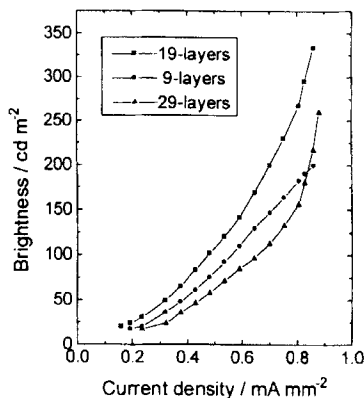


FIGURE 2 L-I characteristics for different layer LaL_2Cl LB films devices.

layer LB films, the electrons injected from the Al electrode were quickly passed through the recombination zone; and in the EL device with 29-layer LB films, the number of electrons that reached recombination zone was decreased because of their energy loss caused by the long-distance transporting procedure. That is, the γ and n_e in both the causes were decreased. Only the EL device with (15-19)-layer LB films has the possibility that not only γ but also n_e increases. Thus, the EL intensity could be enhanced according to eq.(1). That is, the EL devices with (15~19)-layer LB films of LaL_2Cl have the strongest luminance.

A stable greenish-yellow emission was achieved. The wavelength (515 nm) of the emission for the 19-layer LB films was determined at a current density of 0.8 mA/mm^2 . Comparison of the EL emission spectrum of the device with the PL spectrum of the same layers showed that the spectra do not greatly change. The PL and EL peak wavelengths and the half values of these two spectra were 515, 520 nm and 170, 200 nm, respectively. The EL emission spectrum was independent of the driving voltage and current. This result further indicated that the radiative recombination of injected electrons and holes took place in the LaL_2Cl LB films.^[1]

When LaL_2Cl LB films were used as the emitter in EL devices, the deposited surface pressure of the LB films strongly influenced the I-V characteristics and the electroluminescence of the organic EL diode. Fig. 3 shows the I-V characteristics of EL devices with 19-layer LB films deposited at a surface pressures of 8, 20 and 30 mN/m, respectively. When the LB films were prepared at surface pressure of 20 mN/m, the current density crossing this kind

of diodes had stronger nonlinear I-V characteristics and the organic diode had higher EL intensity than another diode prepared at lower surface pressure (8 mN/m). At lower surface pressure, there may be some point defects or pinholes in the LB film in which the high tunnel current would increase exponentially with the field, leading to an increase in the current density crossing the EL device and a decrease in the recombination probability of electrons and holes at higher voltage. When the surface pressure is higher than 20 mN/m, the I-V characteristics of the devices are similar. It indicates there may be fewer point defects or pinholes in these LB films and the difference in quality of these films are small. That is, the LB films with surface pressures higher than 20 mN/m can be important in raising the LaL_2Cl EL intensity and efficiency. Comparing the performance of the LaL_2Cl LB film with those of the amorphous LaL_2Cl film prepared by spin coating at the same voltage, the luminance for the LaL_2Cl LB films deposited at 30 mN/m is about 1.9 times stronger than that for the amorphous LaL_2Cl films of the same thickness (ca. 47.5 nm) prepared by spin coating. The EL spectra are the similar each other. It indicates that the ordered arrangement of the molecules can enhance the emission efficiency of luminescent material.

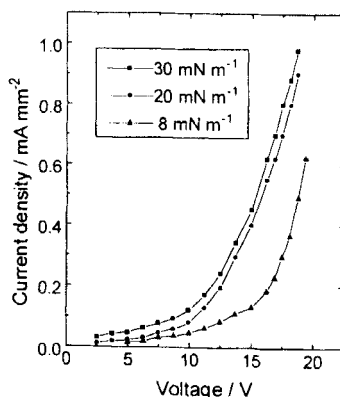


FIGURE 3 Current-voltage (I-V) characteristics for different surface pressure LaL_2Cl LB films devices.

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