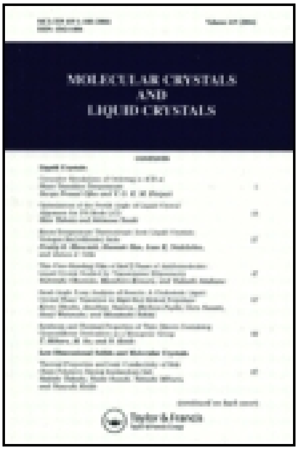

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Improvement of Electrode-Organic Interface by Insertion of Mono-layer Size Al_2O_3 Layer in Organic EL Device

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Improvement of electrode/organic interface has been studied in organic electroluminescent diode which consists of 8-hydroxyquinoline aluminum and diamine derivative. Mono-layer size of aluminum oxide (Al_2O_3) layer inserted between ele-ctrode and organic interface has been studied. Emission efficiency has been increased by inserting mono-layer size of Al_2O_3 between anode and organic layer and between cathode and organic layer. The mechanism of improvement in emission efficiency has been discussed.

Keywords: organic electroluminescent device, metal-organic interface, 8-hydroxyquinoline aluminum, emission efficiency

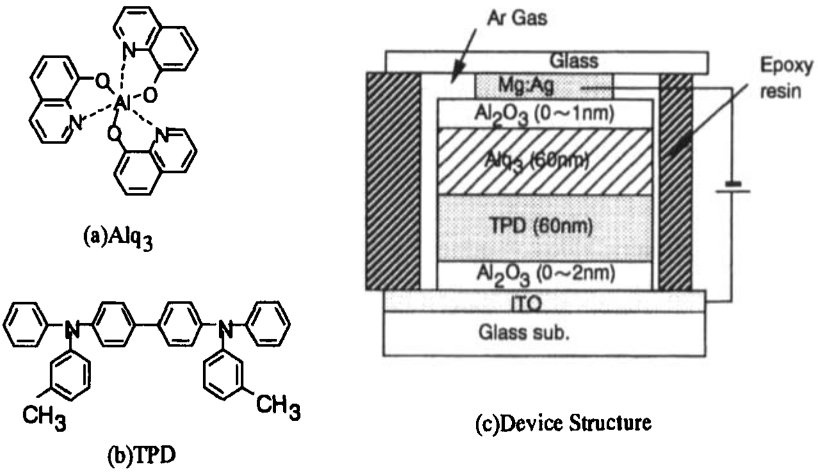
INTRODUCTION

Recently, organic electroluminescent (EL) diodes have studied by many research workers[1-9] to obtain the EL diodes with high efficient, high brightness or with long life time. For improvement of emissive efficiency, the insertion of buffer layer[10,11] is one of the efficient tools. In this study, we examined an effect of insertion of aluminum oxide (Al_2O_3) buffer layer between the ITO anode and the hole transporting layer and between the Mg:Ag cathode metal and the emissive layer in the organic EL diode with diamine derivative for the hole transporting layer and 8-hydroxy-quinoline aluminum (Alq_3) for the emissive layer. The mechanism of enhancement of emission efficiency has been discussed.

EXPERIMENTAL

Figure 1 shows the molecular structure of the organic materials and a schematic de-scription of the EL device used in this study. The device consists of an indium-tin-oxide (ITO)-coated glass substrate, aluminum oxide (Al_2O_3) buffer layer (0-2nm), N, N'-diphenyl-N, N'-(3-methylphenyl)-1,1'-biphenyl- 4,4'-diamine (TPD) hole transporting layer (60nm), 8- hydroxyquinoline aluminum (Alq_3) emissive layer (60nm) and a magnesium containing silver (Mg:Ag) cathode electrode. And we also fabricated the device with Al_2O_3 layer (0-1nm) inserted between the emissive layer and the cathode.

Organic thin films were fabricated using the organic molecular beam deposition (OMBD) system. The deposition was carried out at a back ground



(a)Alq_3

(b)TPD

(c)Device Structure

FIGURE 1 Schematic description of molecular structures and a device structure of Al_2O_3 inserted EL device.

pressure of under 10^{-5} Pa. Organic materials were loaded into separate Knudsen cells (K-cells) and subsequently heated to their sublimation temperature, and then deposited onto the substrate. The layer thickness of the deposited material was monitored in situ using an oscillating quartz thickness monitor. Al_2O_3 layer was fabricated by the method as follows. After deposition of aluminum (Al) on the ITO or organic layer at a back-ground pressure of about 10^{-5} Pa, the device was put in air for about 10 minutes to form Al_2O_3 layer by oxidization of Al layer. The formation of Al_2O_3 layer by the oxidation method was characterized and reported by Li *et al.*[12] Finally Mg:Ag cathode was vapor deposited at a background pressure of about 10^{-5} Pa onto the organic films. After fabricated the layer structures, the device was covered by glass plate and encapsulated by epoxy resin in argon gas atmosphere to prevent the cathode from oxidation. The active device area was 4 mm^2. The forward bias condition is defined as the case in which the ITO electrode is positively biased against the Mg:Ag electrode, and the negative bias vice versa. All the measurements were carried out at room temperature.

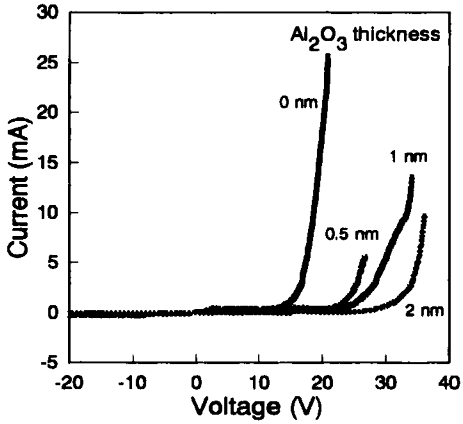
The surface images of glass/ITO surface were carried out by using atomic force microscope (AFM) (JEOL, SPM 4200A).

RESULTS AND DISCUSSION

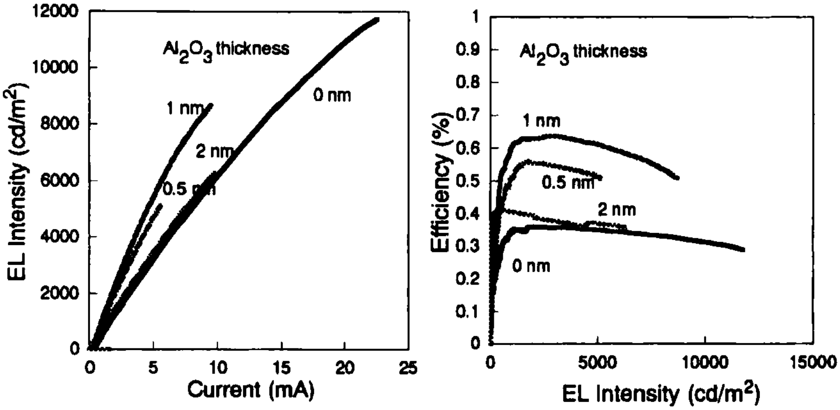
Figure 2(a) shows the applied voltage dependence of injection current characteristics of the Al_2O_3 thin film inserted bet-ween ITO and TPD layers. In the forward biased conditions, the current increased superlinearly with increasing applied voltage, and the current remains in a small value in the reverse bias case. As increas-ing the thickness of aluminum oxide layer the current decreases at the same applied voltage. In the case of no Al_2O_3 layer, the injection current steeply increased at above 15V. However, the injection current in-creases at above 25V in the case of 1nm-thick-Al_2O_3 layer inserted between ITO and TPD. The threshold voltage in the voltage-current characteristics shifted to higher voltage by about 10V by insertion of 1 nm-thick-Al_2O_3 layer.

Figure 2(b) shows the injection current dependence of EL intensity characteristics in the same devices shown in Fig. 2(a). Comparing the emission intensity at same driving current, EL intensity from the devices with 0.5nm-thick-Al_2O_3 or 1nm-thick-Al_2O_3 inserted between ITO and TPD show higher than that of the device with no Al_2O_3 layer.

Quantum efficiency dependence on injection current calculated from the ratio of emitted photons against injected carriers is shown in Fig. 2(c). Quantum efficiency is about 0.35% in the diode with-out Al_2O_3 layer, whereas that about 0.65% in the diode with 1nm-thick-Al_2O_3 layer inserted between the ITO and TPD.



(a)



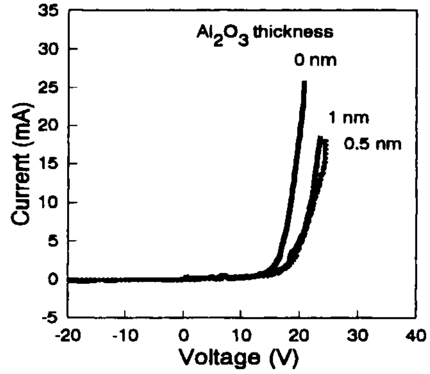
(b) (c)

FIGURE 2 Current vs. applied voltage (a), current vs. EL intensity (b), EL intensity vs. quantum eficiency (c) characteristics of EL device with Al_2O_3 layer inserted between ITO and TPD interface.

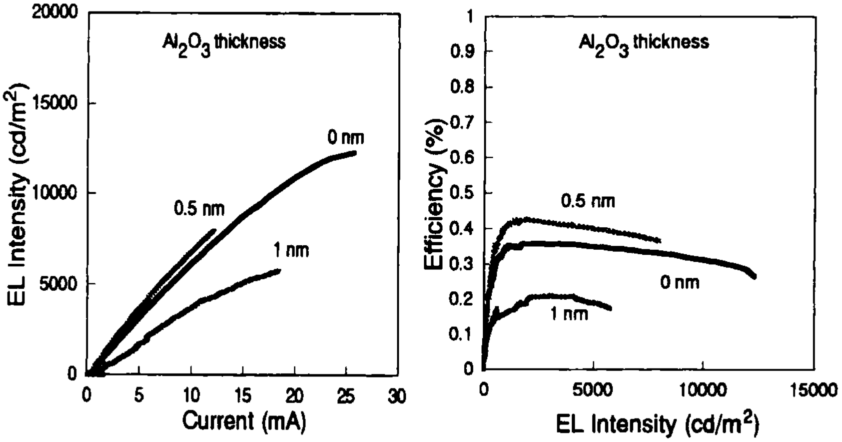
As shown in Fig. 2(c), quantum effi-ciency is the highest among the devices in which 1nm-thick-Al_2O_3 inserted bet-ween ITO and TPD.

Insertion of Al_2O_3 between Alq_3 emissive layer and Mg:Ag cathode has been discussed. Fig. 3(a) shows the injection current dependence on applied voltage characteristics of the device in which Al_2O_3 layer with 0nm, 0.5nm, 1nm inserted between Alq_3 and Mg: Ag cathode.

In the forward bias condition, the current increased super-linearly with increasing the applied voltage, and the current remains in a small value in the negative bias case, which is



(a)



(b) (c)

FIGURE 3 Current vs. applied voltage (a), current vs. EL intensity (b), EL intensity vs. quantm efficiency (c) characteristics of EL device with Al_2O_3 layer inserted between the Alq_3 emissive layer and the Mg:Ag cathode interface.

similar to the case of the device with Al_2O_3 layer inserted between ITO and TPD. As increasing the thickness of Al_2O_3 layer, the injection current decreased under the same applied voltage.

Figure 3(b) shows the injection current dependence of EL intensity characteristics of the devices shown in Fig. 3(a). Under the same driving current, the EL intensity of the diode with 0.5 nm-thick-Al_2O_3 is the highest among the diodes with and without Al_2O_3 layers.

Figure 3(c) shows the injection current dependence on quantum efficiency character-istics of the devices. Quantum efficiency is the highest in the device with 0.5 nm-thick-Al_2O_3 among the devices with various thickness of Al_2O_3 layer of inserted bet-ween the Alq_3 emissive layer and the Mg:Ag cathode.

The mechanism of the emission enhancement by inserting Al_2O_3 layer between electrode and organic layers is explained as follows. First we will discuss the mecha-nism of the carrier transport and the emission mechanism in the EL device with and without Al_2O_3 layer inserted between ITO and TPD layers.

The surface of ITO has about 50 nm roughness by the AFM image. The layer thickness of Al_2O_3 layers is in the range of 0-2 nm inserted between the ITO anodeand the TPD hole transporting layer and that of 0-1 nm inserted between the Alq_3 emissive layer and the Mg:Ag cathode. The Al_2O_3 layers deposited on the ITO or the organic layer are not sufficient enough to cover the whole area, because the roughness of the substrate has much higher roughness than the A_2O_3 layer.

Firstly, the deposited Al_2O_3 layer covers a part of the ITO or the organic layer and is effective for the suppression of the current flow which does not act for the emission. Since the current flow decreases in the diode with Al_2O_3 layer inserted between ITO and TPD layers compared with that without Al_2O_3 layer. The insertion of Al_2O_3 layer depresses the current flow which is not effective for radiative recombination and to decrease the emission efficiency. From the result of current-voltage characteristics that the current-emission characteristics of the diode with Al_2O_3 layer inserted between ITO and TPD, the emission intensity is higher in the diode with Al_2O_3 layer inserted between ITO and TPD,

Secondly, the insertion of the Al_2O_3 layer between the electrode and the organic layer will modulate the energy level at the interface of the organic layer and the metal electrode, since the energy level at the interface of organic layer is reported to be modulated by the metal electrode[13]. The change of energy level at the interface will modulate the current flow at the metal-organic interface, and will change the carrier recombination process.

At this stage of experiment, we cannot conclude which mechanism will be the main process for the improvement of the emission efficeincy, however, we can say that these mechanisms will be one of the reasons for the increase of emission efficiency.

The increase of the insertion of Al_2O_3 layer between Alq_3 and Mg:In electrode will be discussed in the same manner as discussed above. However, the increase in emission efficiency is not so high as that in the case of insertion between the ITO and TPD layers.

We can conclude that the monolayer size of Al_2O_3 layer inserted between electrode and organic layer contribute to enhance the emission efficiency.

COCLUSIONS

The insertion of monolayer size of Al_2O_3 layer having an appropriate layer thickness between the ITO anode and the TPD hole transporting layer, or between the Mg:Ag cathode and the Alq_3 emissive layer improves the emission efficiency of the EL devices.

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