Electrical Conduction Mechanism in ITO/Alq_3/Al Organic Light-emitting Diodes

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We have used ITO/Alq_3/Al structure to study electrical conduction mechanism in organic light-emitting diodes. Current-voltage-luminance characteristics were measured at room temperature by varying the thickness of Alq_3 layer from 60 to 400mm. We were able to confirm that there are three different mechanisms depending on the applied voltage region; ohmic, space-charge-limited current, and trap-charge-limit-current mechanism. And the maximum luminous efficiency was obtained when the thickness of Alq_3 layer is 200nm.

Keywords : Organic light-emitting diodes, Conduction mechanism, Luminous efficiency, Thickness

1. INTRODUCTION

There is a growing concern in organic light-emitting diodes since a report of organic luminescent phenomena in 1963 by Pope using anthracene single crystals[1]. In 1987, Tang and VanSlyke in Eastman Kodak observed green light emission at low voltage using low molecule aromatic diamine and 8-hydroxyquinolinato aluminum (Alq_3)[2]. In 1990, Friend et al., in Combridge University reported the first green light-emitting polymer diodes using poly(phenylenevinylene) (PPV).

These kinds of organic light-emitting diodes are attractive because of potential application in display in the near future. As a living and office space becomes expensive, there is a need to use an interior space more effectively. Thus, a thin, light, and high-quality displays are need, which occupy less space. One of the displays satisfying these requirements is an organic light-emitting diodes. To use this device more effectively, it is important to understand a conduction mechanism in organic light-emitting diodes.

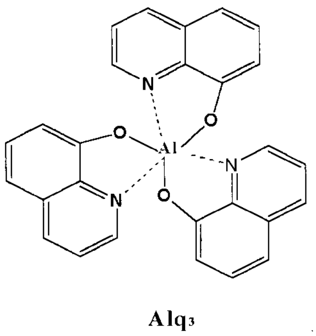
Ma Dongge used Mott-Gurney equation and energy-band model to explain the current-voltage characteristics[3]. Parker studied a current-voltage characteristics in ITO/MEH-PPV/cathode structure by varying cathodes (In, Al, Ag, Cu and Au)[4]. He classified majority and minor carriers depending on energy barrier between cathode and MEH-PPV, and explained the electrical conduction phenomena using a tunneling model. And Lee et al., used a tunneling model to explain the conduction mechanism in polymer light emitting diodes of poly(p-phenylene)[5].

The Alq_3 organic material is widely used for emissive and electron-transport layer[6]. In this paper we report electrical conduction phenomena in organic light-emitting diodes through a study of current-voltage-luminance characteristics depending on a layer thickness of Alq_3.

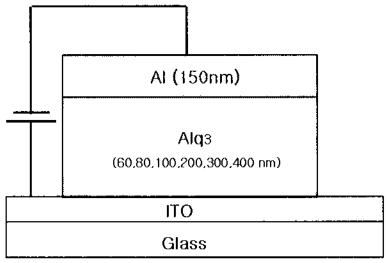
2. EXPERIMENTAL

To study a conduction mechanism using Alq_3 layers, we fabricated a device structure of ITO/Alq_3/Al. Figure 1(a) is a molecular structure of Alq_3 and Fig. 1(b) is a schematic representation of device structure. Current-voltage-luminance characteristics were analyzed in a device structure of ITO/Alq_3/Al by varying a layer thickness of Alq_3 (60, 80, 100, 200, 300, and 400nm).

Figure 2 shows AFM(Atomic Force Microscope) images of ITO(indium-tin-oxide) substrate surface and Alq_3(100nm) film surface. These measurements show that a surface roughness of ITO is 1.8nm and that of Alq_3 is 1.9nm. The ITO substrate, which was used as an anode, has a surface resistance of 15Ω/□ and thickness of 170nm. Electrical insulating tape having a width of 5mm was attached to the ITO surface to make a patterned ITO line. The taped ITO surface was exposed to the vapor of HCl and HNO_3 made with a volume ratio of 3:1.

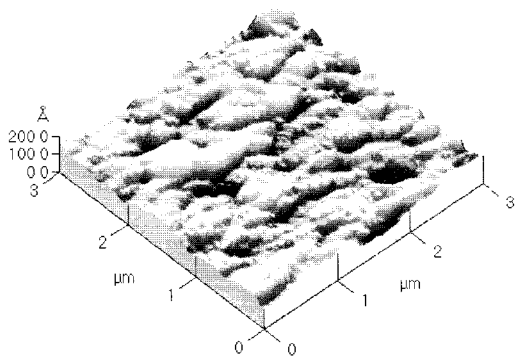


(a) Molecular structure of Alq_3.

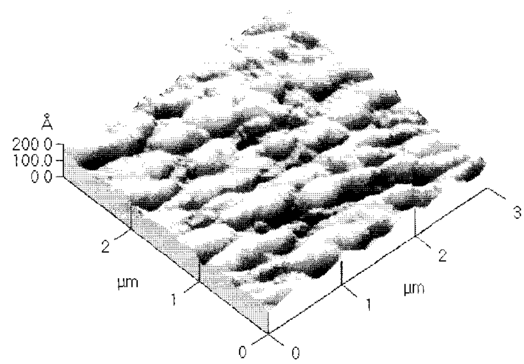


(b) $ITO/Alq_3/Al$ device structure.

Fig. 1. Molecular structure and device structure.



(a) AFM image of ITO surface.



(b) AFM image of Alq_3 (100nm) film.

Fig. 2. AFM images of ITO substrate and Alq_3 (100nm) film surface.

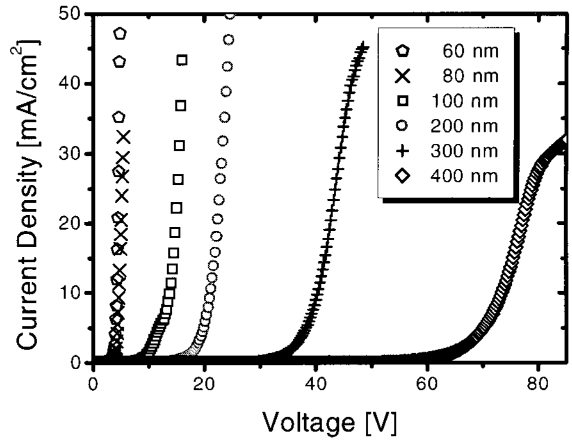


Fig. 3. Current density-voltage characteristics with thickness variation of Alq_3 in $ITO/Alq_3/Al$ device structure.

After removing the tape from the ITO, the ITO surface exposed to the chemical vapor was removed by cleaning it ultrasonically in a chloroform solution for 20 minutes. And then the cleaned ITO was heated to 80℃ in a mixed solution of distilled water, H_2O_2 and ammonia made with a volume ratio of 5:1:1 for 60 minutes. After these treatments, the ITO was ultrasonically cleaned again in chloroform and distilled water for 20 minutes, respectively. And then it was dried with a blow of nitrogen gas.

The Alq_3 purchased from TCI was thermally evaporated onto the ITO surface at a deposition rate of 0.7 Å/s under a base pressure of 5×10^{-6}torr. And then the aluminum of 150nm thickness was thermally evaporated on Alq_3 layer under a base pressure of 5×10^{-6} torr. A deposition rate of aluminum was 0.5 Å/s up to 10nm thick, and 10 Å/s in 10nm~150nm thickness range. A light-emitting area was $15mm^2$($5mm\times 3mm$).

Current-voltage-luminance characteristics were measured at room temperature using Keithley 236 source-measure unit, 617 electrometer, and Si-photodiode (Centronics Co. OSD 100-5T).

3. RESULTS AND DISCUSSION

Figure 3 shows a current density-voltage characteristics in $ITO/Alq_3/Al$ device structure for several thicknesses of Alq_3 layer. As shown in Fig. 3, there is a sharp current density in thin Alq_3 layer. However, as the thickness of Alq_3 layer increases, the current density increases rather slowly. When the thickness of Alq_3 layer is 400nm, the device was survived even when the applied voltage is 100V.

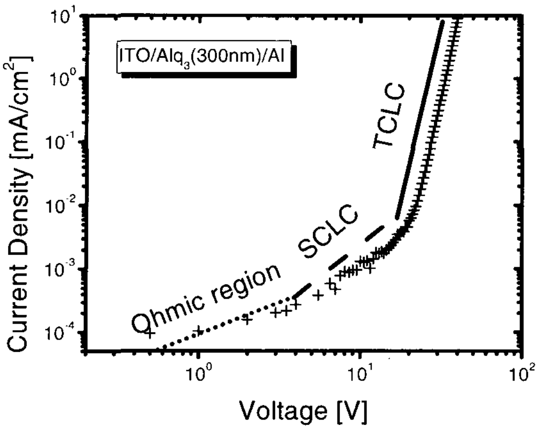


Fig. 4. Current density-voltage characteristics in ITO/Alq_3 (300nm)/Al structure.

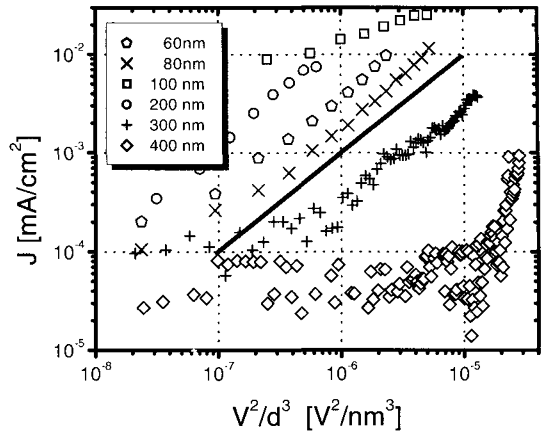


Fig. 5. Current density-voltage characteristics in space-charge-limited-current region.

Figure 4 shows the current density-voltage characteristics to explain the conduction mechanism for Alq_3 layer of 300nm thickness. In the figure, the conduction mechanism can be classified into three regions; ohmic region, space-charge-limited current(SCLC) region, and trap-charge-limited current(TCLC) region depending on the region of applied voltage[7-9].

When the applied voltage is low in ITO/Alq_3(300nm)/Al device, the conduction is dominant by thermally generated residual charge carriers in the bulk rather than the injected charge carriers. In this case, the current density through the Alq_3 layer is governed by Ohm's law. That is

$$J_{ohm} = en_0\mu\frac{V}{d} \tag{1}$$

Here, e is the electric charge, n_0 the charge carrier density, μ the charge carrier mobility, V the applied voltage, and d the thickness of organic layer.

As the applied voltage increases, the injected charge from the electrode to the HOMO(Highest Occupied Molecular Orbital)/LUMO(Lowest Unoccupied Molecular Orbital) forms a space charge near the interface, and thus the current is limited by the space charge. This is called the space-charge-limited current(SCLC). If there is no trap states in SCLC region, a relation between the current density J and the applied voltage V has the following relation.

$$J_{SCLC} = \frac{9}{8}\varepsilon\mu\frac{V^2}{d^3} \tag{2}$$

In reality, there exist traps in the organic layer, so that it is not simply described by the single discrete energy level. If traps are not filled by charge carriers, the current is reduced due to a trap of charge carriers. Then Eq. (2) can be modified into the following, such as

$$J_{SCLC} = \frac{9}{8}\varepsilon\mu\theta\frac{V^2}{d^3} \tag{3}$$

Equations (2) and (3) are called Mott and Gurney formula. Here,

$$\theta = \frac{n}{n+n_t} \quad \text{or} \quad \theta = \frac{p}{p+p_t} \tag{4}$$

where, θ is a fraction of free carriers relative to the total number of carriers; n is a free electron density, n_t a trapped electron density, p a free hole density, and p_t a trapped hole density. Kusano et al., mainly exploited the space-charge-limited current before studying tunneling behavior. From Eqs. (2) and (3), the space-charge-limited-current region satisfies a slope of 1 in a plot of log(J) vs log(V^2/d^3).

Figure 5 shows a plot of log(J) vs log(V^2/d^3) for several thicknesses of Alq_3 layer to see a space-charge-limited-current mechanism. A solid line in the figure represents a slope of 1. We can see that there exists a region with slope of 1 regardless of the Alq_3 layer thicknesses. It indicates that the space-charge-limited current flows before the light emission occurs in the device. In Fig. 5, the slope is much less than 1 for the device with 400nm thick. This low value of slope comes from the low value of current. This low value of current is not reliable because our electrometer has a limit of current measurement.

If a trap distribution is exponential in the forbidden band such as

$$h(E) = \frac{N_t}{E_c}\exp(-\frac{E}{E_c}) \tag{5}$$

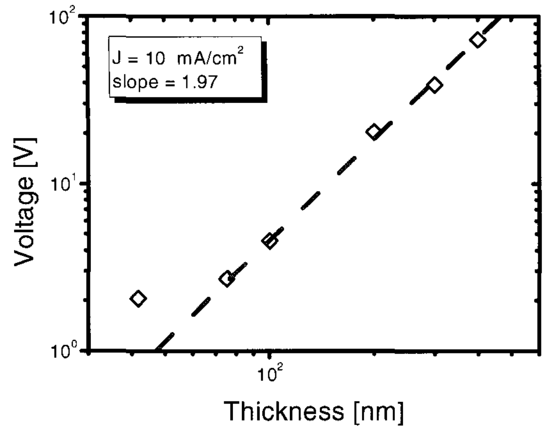


Fig. 6. Thickness-dependent voltage under the constant current density of 10mA/cm^2 in ITO/Alq$_3$/Al device.

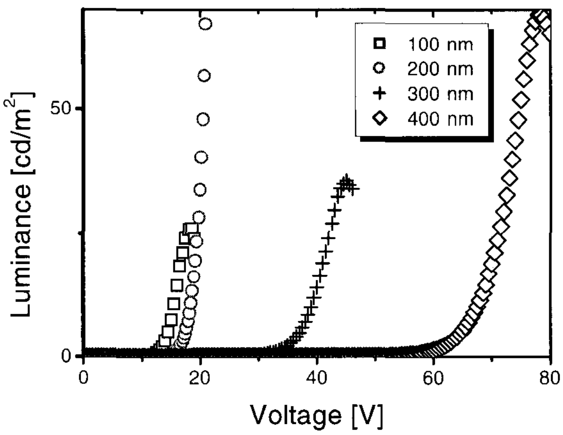


Fig. 7. Luminescence-voltage characteristics with thickness variation of Alq$_3$ in ITO/Alq$_3$/Al device.

where, N_t is a trap density in the center and E_c is a characteristic energy of the trap distribution. The following formula is a relation of current density-voltage characteristics in this trap-charge-limited-current region.

$$J \propto \frac{V^{m+1}}{d^{2m+1}} \tag{6}$$

A voltage region satisfying Eq. (6) gives a light emission in the device. In Eq. (6), the value of m is usually greater than 6. If m is large, a relation between log(V) and log(d) gives a slope of 2 under a constant current density. Figure 6 shows a plot of log(V) vs log (d) under the constant current density of 10mA/cm^2. The slope of dotted line in the figure is 1.97, which is close to the expected value of 2. Thus, there exists a conduction due to trap-charge-limit current in the high voltage region.

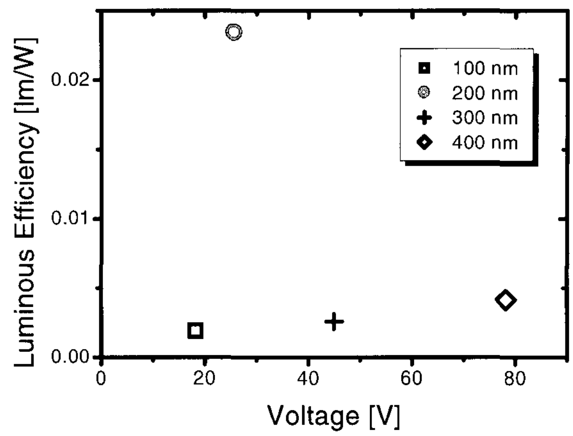


Fig. 8. Maximum luminous efficiency-voltage characteristics with thickness variation.

Figure 7 shows a luminance-voltage characteristics in ITO/Alq$_3$/Al device for several thicknesses of Alq$_3$ layer. As the layer thicknesses increases, the operating voltage increases as well. A maximum luminance was obtained when the Alq$_3$ layer thickness is 200nm.

Using the current-voltage-luminance characteristics of the device, a luminous efficiency of device was calculated. Figure 8 shows the maximum luminous efficiency as a function of the applied voltage for the several thicknesses of Alq$_3$ layer. The maximum luminous efficiency was obtained to be 0.023lm/W for 200nm thick Alq$_3$ layer. And the efficiencies are lower for the other Alq$_3$ layer thicknesses.

4. CONCLUSION

An electrical conduction mechanism was studied in the device structure of ITO/Alq$_3$/Al by varying the thickness of Alq$_3$ layer. We were able to classify the conduction mechanism in organic light-emitting diodes into three different regimes depending on the region of applied voltage; ohmic region, space-charge-limited-current region, and trap-charge-limited-current region. And the maximum luminous efficiency of 0.023lm/W was obtained for 200nm thick Alq$_3$ layer.

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