Frequency Dependent Properties of Tris(8-Hydroxyquinoline) Aluminum Thin Films

Yong-Soo Lee, Jae-Hoon Park and Jong Sun Choi

Abstract - Admittance or impedance spectroscopy is one of the powerful tools to study dielectric relaxation and loss processes in organic and inorganic materials. In this study, the frequency dependent properties of an indium tin oxide/tris(8-hydroxyquinoline) aluminum(Alq$_3$)/aluminum structure have been studied. The conductance of the Alq$_3$ film increases with the DC applied voltage up to 4V and decreases above 4V in the low frequency region. This indicates that the resistance of the device decreases with the applied bias due to the carrier injection enhancement, thereafter the injected carriers form the space charge and the additional injection of carriers is prevented. The Cole-Cole plot of the admittance takes a one-semicircle shape, which means that the device can be modeled as a parallel resistor-capacitor network. The resistance and capacitance were estimated as 8.62kΩ and 2.7nF, respectively, at 3V in the low frequency region. The dielectric constant (ε') of the Alq$_3$ film is independent of the frequency in the low frequency region below 100kHz, while the frequency dependency was observed at above 100kHz. The dielectric loss factor (ε'') of the Alq$_3$ film shows the dielectric dispersion below 100kHz and dielectric absorption in higher frequency domain. The dispersion is thought to be related to the hopping process of the carriers. The ε'' is proportional to the reciprocal of the frequency. The dielectric relaxation time was extracted to be about 0.318μs from the dielectric absorption spectrum.

Keywords - organic light emitting diode, admittance, frequency dependence, dielectric constant, dielectric loss factor

1. Introduction

Some organic materials have received considerable attention as semiconductors for the device applications such as light-emitting diodes, thin film transistors, and solar cells [1-3]. The organic light emitting diodes based on small molecules or conjugated polymers have been of greater interest due to their possible applications for flat panel displays. They are attractive because of the capability of multicolor emission, low operating voltage, and competitive cost. A lot of investigations for commercial applications has been pursued, but some of the important electrical characteristics have not been revealed yet. The electrical characteristics of the interfaces between organic layer and metal electrodes, and between organic layers are still not clearly understood. Admittance or impedance spectroscopy is one of the powerful tools to study equivalent circuit models, and relaxation and loss processes in organic and inorganic materials and devices [4-6]. In this study, we have investigated the single layer organic light emitting diodes with an indium tin oxide (ITO) / tris(8-hydroxyquinoline) aluminum (Alq$_3$) / aluminum structure. The frequency dependent properties and the equivalent circuit models of the Alq$_3$ thin film devices are presented.

2. Experimental

Glass substrates were ultrasonically cleaned with acetone, isopropyl alcohol, and deionized water, and then dried in a vacuum oven. The devices were fabricated on the ITO-patterned glass substrates. ITO has a sheet resistance less than 20$\Omega/\square$ and is about 100nm thick. The organic material was thermally evaporated on the patterned ITO, under 2×10^{-6}Torr. An aluminum cathode was evaporated under the same vacuum condition. During the evaporation, the substrates were held at room temperature. The effective cell area is defined with the overlap region between the anode and the cathode, as 0.09cm^2. The thicknesses of the organic layer and cathode were about 120nm and 100nm, respectively. The thickness is confirmed using ellipsometry (Plasmos, SD-2100) and an α-step profilemeter (Tenkor, 200). The positive bias is applied to ITO with respect to aluminum. Frequency dependent properties were measured by an HP 4192A LF impedance analyzer from 100Hz to 10MHz. The AC oscillating signal is superimposed on the operating DC bias. The amplitude of the

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oscillating signal was 50mV. The AC current is out of phase with AC voltage and complex relationship is as follows:

$$Y=\frac{1}{Z}=G+jB=G+j\omega C \qquad (1)$$

where Y is admittance, Z impedance, G conductance, B susceptance, and C capacitance, respectively. All the measurements were carried out in the dark and shielded environment. The inset of Fig. 2 shows the molecular structure of Alq_3.

3. Results and Discussion

Fig. 1 shows the conductance of the single layer device (area: $0.09cm^2$, thickness: 120nm) as a function of the frequency and applied DC bias. The inset is the conductance variation with the applied voltage. The applied voltages are 0, 2, 3, 4, 5, and 6V. The measurable values can be obtained from 3V. In the low frequency region, the bias dependent properties of conductance are observed, and the conductance is independent of the bias at higher frequencies. The conductance of the film increases with the applied voltage up to 4V and decreases above 4V in the low frequency region. This indicates that the resistance of the device decreases with the applied bias due to the carrier injection enhancement, thereafter the injected carriers form the space charge at the organic bulk or interface between the organic layer and the electrode, and the additional injection of the carriers is prevented. The resistance at low frequencies is about $8.62k\Omega$ at 3V, which is the resistance of the Alq_3 bulk region, and the corresponding electrical conductivity of the device is around 1.55×10^{-8}S/cm. Fig. 2 shows the measured capacitances with varying the frequency and bias. The capacitance of the Alq_3 film exhibits the dispersion properties under all the measured frequencies, which are thought to be related to the localized trap charges in the bandgap of the organic material. The capacitance is independent of the bias above 500kHz. In the low frequency domain, the capacitance slightly decreases up to 4V, and then increases above 4V, which might indicate a space charge formation. This configuration shows quite similar tendency to the conductance data. The capacitance is 2.7nF ($30nF/cm^2$) at 3V, in the low frequency region.

Fig. 3 shows the Cole-Cole plot of the single layer device. The horizontal axis is the real part of the admittance and the vertical axis the imaginary part of the admittance of the Alq_3 film. The frequency increases from left to right along the horizontal axis. The admittances in the low frequency region is dependent on the bias, whereas in the high frequency region the data are more or less bias independent. The plot takes a one-semicircle shape, which means that the device can be modeled as an equivalent circuit with a parallel resistor-capacitor network. The same result can be obtained for the thinner device. The measurement of the Alq_3 single layer device with the magnesium cathode was performed by Ono et al. [7]. They measured the impedance of the device in vacuum and air environments. Their results in the vacuum condition are similar to our experimental results, though we measured in air condition. But, their results measured in the air are different from ours. They suggested the two circuits connected in series, each of which is composed of the parallel resistor-capacitor circuit, as an equivalent one to reflect the formation of the additional layer at the interface between Alq_3 film and magnesium electrode. This is due to the diffusion of magnesium into the Alq_3 layer in the air. Therefore, the magnesium doped Alq_3 layer is believed to be formed at the organic layer/

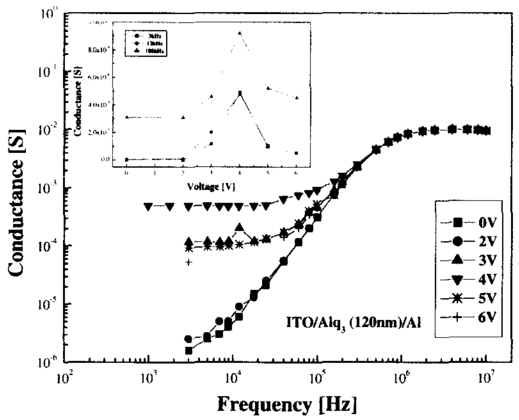

Fig. 1 Conductance of the device as a function of frequency and an applied bias; inset, the conductance with the applied bias.

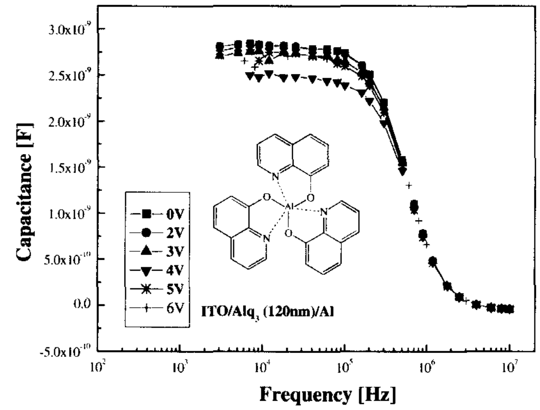

Fig. 2 Capacitance with frequency and a DC bias; inset, the molecular structure of Alq_3.

magnesium electrode interface. Consequently, we assumed that the additional layer at the interface between Alq$_3$ film and aluminum cathode is not formed in our experiment [8].

The inset of Fig. 3 shows the equivalent circuit of the single layer device. R$_P$ and C$_P$ are the Alq$_3$ bulk resistance and capacitance, respectively, and R$_S$ is the series resistance at the anode/Alq$_3$ contact.

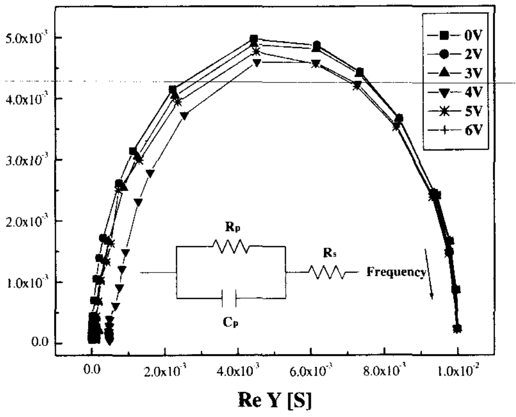

Fig. 3 Cole-Cole plot of the Alq$_3$ single layer device; inset, the equivalent circuit of the device.

Fig. 4 shows the conductance of the device as a function of frequency, which is obtained by an equivalent circuit simulation. The equivalent circuit is composed of one parallel resistor-capacitor network in series with a resistor. The simulation was performed at 3V DC bias, from 100Hz to 10MHz frequency range. Series resistance (R$_S$) is about 100Ω, which is much less than bulk resistance. The result is in very good agreement with Fig. 1, and the theoretical equivalent circuit could describe the experimental result quite well.

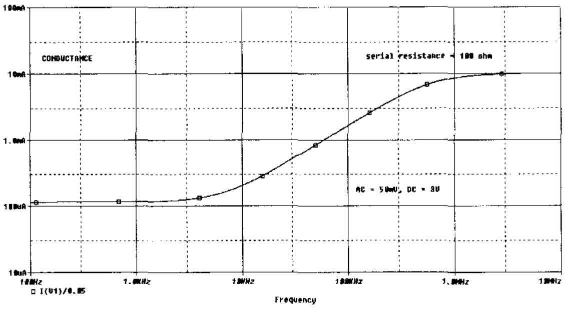

Fig. 4 Equivalent circuit simulation result for the single layer device.

The complex permittivity describes the charge storage capability and the loss process in the films. Fig. 5 shows the real part of the complex permittivity (ε') as a function of frequency and an external bias. The real part of the complex permit-tivities is related to the capacitances, as shown in Fig. 2, by the equation below:

$$\varepsilon' = \frac{d}{\varepsilon_0 A} C \qquad (2)$$

where ε' is the real part of the complex permittivity, ε_0 the permittivity of the free space, A the effective area, d the thickness of the film, and C the capacitance of the material, respectively. The permittivity in the low frequency region is about 4.1 at 0V application.

Fig. 6 shows the variation of the imaginary part of the permittivity (ε'', loss factor) with the frequency and bias. In the low frequency domain, the bias dependent properties were observed. The bias independent properties are measured at higher frequencies. The loss factor shows the dielectric dispersion below about 100kHz and dielectric absorption in the higher frequency region. The dispersion is thought to be related to the hopping process of the carriers, and the loss factor is proportional to the reciprocal of the frequency [9]. These results are similar to those of Kim et al. [10]. They reported the frequency dependent properties of the imaginary part of the capacitance for MEH-PPV single layer light emitting diodes. In the low frequency region, the charge carriers more alternately and slowly enough to continuously hop over the potential barriers and, therefore, make DC conductivity [10]. The loss factor shows a peak at around 500kHz as can be seen in Fig. 6. That is related to the dipolar polarization [11] and the corresponding dielectric relaxation time is extracted to be about 0.318μs, which determines the mean time for the dipole to lose its alignment.

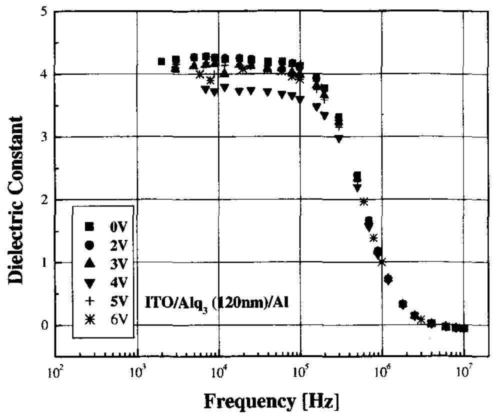

Fig. 5 Real part of the complex permittivity as a function of frequency and a bias.

Fig. 7 shows the variation of the imaginary part of the complex permittivity with the thickness of the Alq$_3$ film. The maximum frequency in the high frequency region, which is related to the dielectric relaxation, is constant with the thickness. This indi-cates that the thickness variation, namely the change of the electric field (E=V/d), doesn't influence the relaxation of the electric dipole in the Alq$_3$ film. In the dielectric dispersion, even with the thickness variation, the dielectric loss factor is proportional to the reciprocal of the frequency.

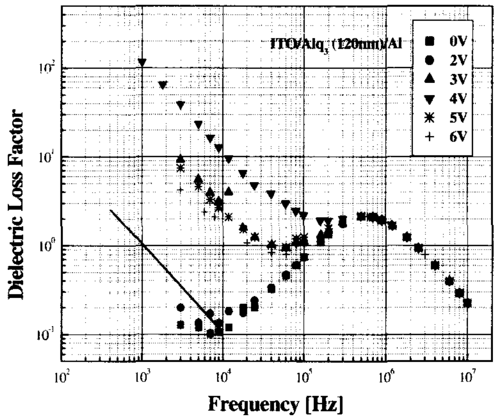

Fig. 6 Imaginary part of the permittivity with frequency and a bias.

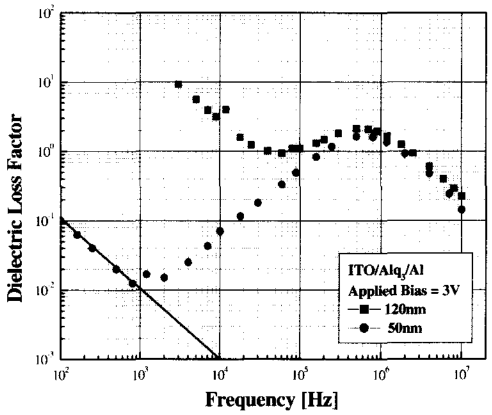

Fig. 7 Imaginary part of the permittivity with thickness.

4. Conclusion

The frequency dependent properties of indium tin oxide/tris(8-hydroxyquinoline) aluminum(Alq$_3$)/aluminum devices have been investigated. The resistance of the organic layer bulk decreases with the applied bias due to the carrier injection enhancement, thereafter the injected carriers form the space charge and additional injection of the carriers is prevented. The Cole-Cole plot of the admittances takes a one-semicircle shape, which means that the device can be modeled as a simple parallel resistor-capacitor equivalent circuit. The real part of the permittivity of the Alq$_3$ film shows frequency independence in the low frequency region below 100kHz, while the frequency dependency was observed at above 100kHz. The imaginary part of the permittivity shows the dielectric dispersion below 100kHz and the dielectric absorption in the higher frequency domain. The dispersion is thought to be related to the hopping process of the carriers, and the loss factor is proportional to the reciprocal of the frequency. The dielectric relaxation time was extracted to be about 0.318 μs from the dielectric absorption spectrum.

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