Journal of Ceramic Processing Research. Vol. 13, Special. 2, pp. s163~s165 (2012)

J O U R N A L O F
Ceramic Processing Research

Temperature-dependent dielectric relaxation in ITO/Alq_3/Al organic light-emitting diodes

Joonho Ahn[a], Tae Wan Kim[b] and Won Jae Lee[c,]*

[a]*Electric Industry Research Institute of Korea, Seoul, 157-718, Korea*
[b]*Department of Physics, Hongik University, Seoul, 121-791, Korea*
[c]*Department of Electronic Engineering, Gachon University, Kyunggi-Do, 461-701, Korea*

Impedance spectroscopy informs electrical properties of materials as accumulated charges, contact status between electrode and organic materials. We carried out impedance spectroscopy of organic light-emitting diodes as ITO/Alq_3(60 nm)/Al on temperatures from 10 K to 300 K. The result described Z'-Z" plot, cole-cole plot and dielectric relaxation time τ. Z'-Z" plot means that real and imaginary part of materials in organic and electrode by frequencies and temperature. Z' as real part of impedance by applied frequency depending on temperature shows the plateau in low frequency region as $R_s + R_p$ and over 100 kHz in high frequency region as R_s. Cole-cole plot shows resistance of materials in equivalent circuit of the device by temperatures. And equivalent circuit and dielectric relaxation could be accomplished by using the complex impedance analysis.

Keywords: Impedance, Organic light-emitting diodes, Temperatures.

Introduction

Organic light-emitting diodes expanding fields widely using flat panel display as well as like organic TFT due to have many advantages that low applied voltages, wide view angles, fast response and low prices. Therefore, for variable application, it is needed to study not only light emitting efficiency but a mechanism of organic light-emitting diodes. But the mechanism of organic light-emitting diodes has not been fully understood until now.

In 1994, I. D. Parker [1] et al. studied a current-voltage characteristics single layer (ITO/MEH-PPV/cathodes) structure by varying cathodes that In, Al, Ag, Cu, and Au. They classified two parts of majority and minority carrier depending on energy barrier between cathode and organic layer and explained the electrical conduction phenomena using a tunneling model. Jonda [2] investigated the electronic properties of organic light-emitting diodes devices by impedance spectroscopy. He carried out the impedance measurement in single layer and double layer that ITO / PVK + TDAPB-4 + Alq_3(4 : 1 : 1) / Mg : Ag(10 : 1) and ITO / PVK + TDAPB-4(1 : 1) / Alq_3 / Mg:Ag(10 : 1) by frequencies (100 Hz ~ 1 MHz). The results show decreased real part and imaginary part of impedance Z by applied voltages and frequencies and interpreted equivalent circuit analysis consist of series R_s and parallel R_p and C_p. Roy [3] et al. reported the electronic properties in layer-by-layer thin film by impedance characteristics. They aimed at getting an understanding of how the electrodes affect the conductance in different ranges of frequency and how a dc bias voltage causes the saturation of polarization. And they analyzed the organic layer in terms of resistive and capacitive component.

This work presents the study on impedance spectroscopy depending on temperature that shows to change device characteristics by the change of electronic material's properties. The result confirmed that impedance decreased with increasing temperature.

Experimental

The device was fabricated to single layer structure of ITO / Alq_3(60 nm) / Al. A thermal evaporation was employed to evaporate Alq_3 at a base pressure of 5×10^{-6} torr with a deposition rate of 0.3 ~ 0.5 Å/s. And aluminum was also thermally evaporated to a thickness of 100 nm at a base pressure of torr and a deposition rate of aluminum was 0.5 Å/s up to 10 nm thick, 10 Å/s in 10 nm ~ 100 nm thickness range.

A current-voltage characteristic of the devices was measured by using Keithley 236 source-measure unit, Keithley 617 electrometer, and Si-photodiode (Centronics Co. OSD100-5T). A frequency and voltage-dependent response of the device was measured using Agilent 4294 A precision impedance analyzer in the range of 40 Hz ~ 100 MHz. An amplitude of the ac signal was set to 100 mV. And the temperature-dependent impedance characteristics were measured using CTI-

*Corresponding author:
Tel : +82-31-750-5813
Fax: +82-31-750-5813
E-mail: wjlee@gachon.ac.kr

Cryogenics refrigerator (Model 22, 8200 compressor Helix Technology Co.).

The frequency-dependent impedance of the device was measured from 40 Hz ~ 100 MHz at different voltages of -4 V, 0 V, 2 V, 4 V, 20 V in variable temperatures 10 K, 50 K, 100 K, 150 K, 200 K, 250 K, 300 K, respectively. All measurements were carried out in vacuum system.

Results and discussion

A complex impedance Z expressed in terms of real and imaginary component Z' and Z'' such as

$$Z = Z' - jZ'' = |Z|e^{j\theta} \tag{1}$$

Here, $|Z|$ and θ are a magnitude and phase of the impedance, respectively.

We assumed an equivalent circuit that was composed a series R_s and a parallel R_p and C_p. R_s is contact resistance between electrode and organic layer. R_p and C_p is a resistance and capacitance component in the device, respectively [4].

$$Z' = R_s + \frac{R_p}{1+(\omega\tau)^2} \tag{2}$$

$$Z'' = \frac{\omega\tau R_p}{1+(\omega\tau)^2} \quad \text{where} \quad \tau = R_p C_p\,. \tag{3}$$

Fig. 1(a) shows the magnitude of impedance by an applied voltage depending on temperature at 100 Hz.

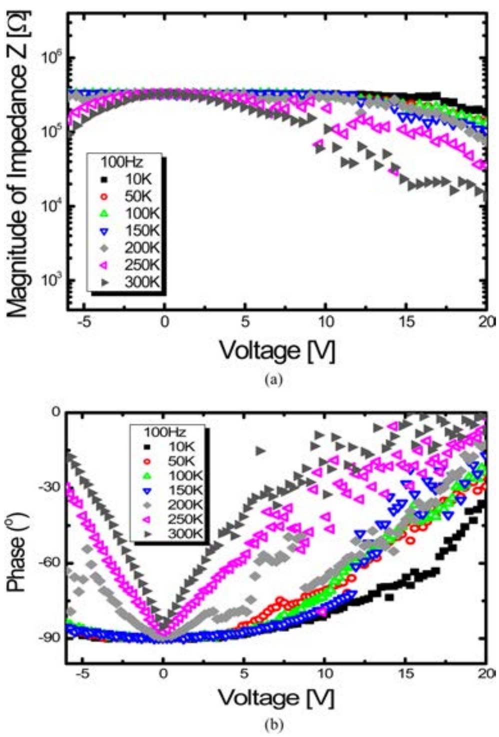

Fig. 1. (a) Magnitude of impedance and (b) corresponding phase of impedance depending on temperature at 100 Hz.

The magnitude of impedance is constant in the measured voltage range for a low temperature. As the temperature increases, the magnitude of impedance decreased by applied voltage. The magnitude of impedance is about 325 kΩ at 300 K.

Fig. 1(b) shows corresponding phase of impedance by applied voltage depending on temperature at 100 Hz. Corresponding phase shows resistance and capacitance component. When phase approaches to 0 °, it exhibited resistance component and it seems to capacitance component on -90 °. Capacitance component in the device means to charge each side as the anode and cathode. Resistance component explain to the resistor in the device. This figure shows it approached to 0 ° at high temperature and voltage.

Fig. 2(a) shows real part of impedance by applied frequency depending on temperature. Magnitude of impedance shows to decreased due to increasing temperature. At 300 K, it is seen the plateau in low frequency that made by resistance $R_s + R_p$ in the device. It was about 9 kΩ~10 kΩ. And this plateau decreased dramatically between 1 kHz and 100 kHz, which is attributed to the capacitance and resistance together. Under 100 kHz, it is seen the second plateau that has resistance R_s in the device.

When the plateau goes to the end of x axis, about 1 MHz, it is seen again that it has no infected capacitance (C_p) and parallel resistance (R_p) and remands only R_s. This value is about 20 Ω. It was similar to find R_s value by Cole-Cole plot. And the

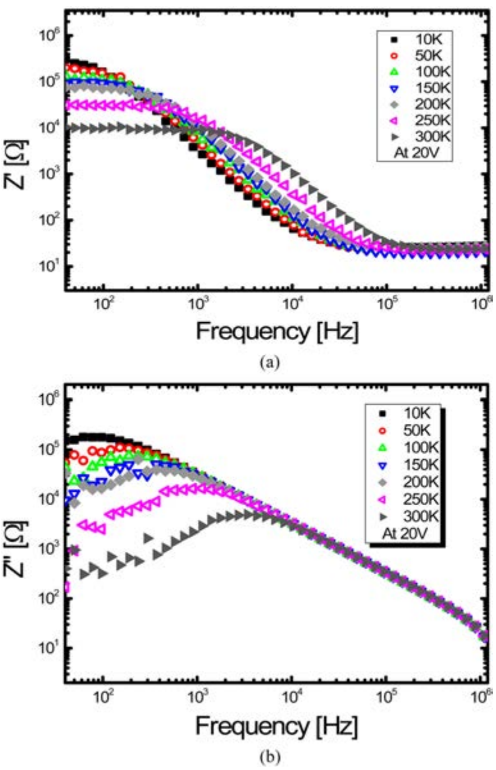

Fig. 2. (a) real part and (b) imaginary part vs frequency at 20 V depending on temperature.

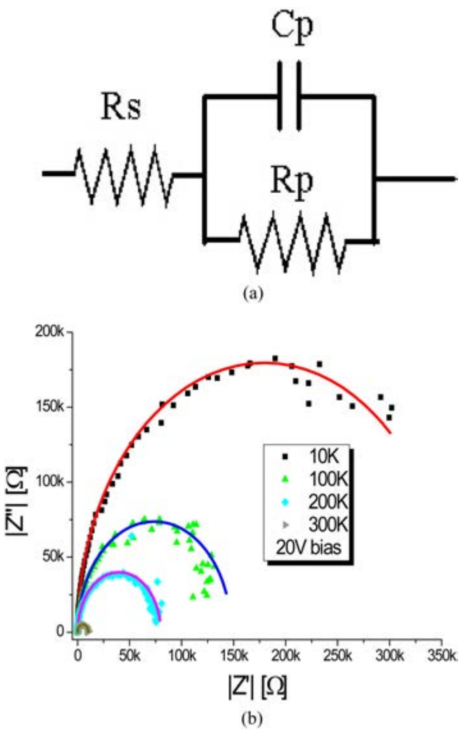

Fig. 3. (a) Equivalent-circuit model in terms of contact resistance R_s in series with parallel combination of resistance R_p and capacitance C_p and (b) Cole-Cole plot under the applied voltage of 20 V.

plateau expanded to the high frequency along to increasing temperatures. Change of plateau means change of resistance and capacitance depending on temperature. At low temperature, the device has high capacitance due to high resistance on the device. And at high temperature, the device has low capacitance due to low resistance by large current.

Fig. 2(b) shows an imaginary part of impedance by applied frequency depending on temperature at 20V. With increasing temperature, peak of Z" decreased and shifted to increasing frequency.

Fig. 3 shows Cole-Cole plot depending on temperature. Cole-Cole plot expressed semicircular following equations,

$$\left[Z'-\left(R_s+\frac{R_p}{2}\right)\right]^2+[Z'']^2=\left[\frac{R_p}{2}\right]^2 \quad (4)$$

In Z' - Z" plane is an equation of circle, having a center at $(R_s+R_p/2,0)$ with radius of $(R_p/2)$. From this analysis, it gives Rs about 20 Ω and the radius gives a resistance R_p around 359 kΩ (10 K), 147 kΩ (100 K), 79 kΩ (200 K), 10 kΩ (300 K) respectively. And C_p was around 4.9 nF.

This circular equation can be expressed alike Cole-Cole semicircular had asserted in the most of dielectric materials. And the dielectric relaxation at low frequency is seen in Fig. 3.

Fig. 4 shows $\tau = C_pR_p$. τ means dielectric relaxation time of device and decreased slightly by temperatures.

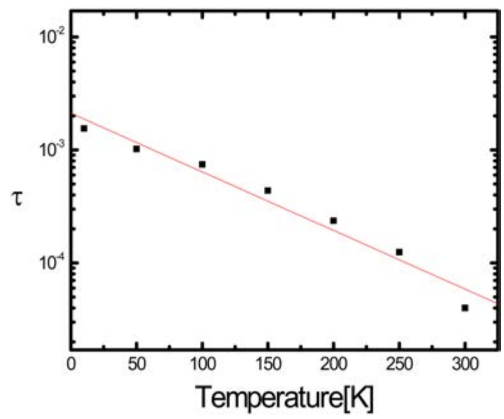

Fig. 4. Dielectric relaxation time $\tau = C_pR_p$ wih increasing temperature at 20 V.

Relaxation time in low temperature seems to be shorter than high temperatures. It was related to the decrease and shift to the right in Fig. 3(b).

Conclusions

This work reported impedance spectroscopy depending on temperature. The magnitude of impedance was constant in the measured voltage range for a low temperature and decreased with increasing voltage and temperature. Phase θ measured to be - 90 o at 0 V in range of 10 K ~ 300 K and is gradually approaches 0 ° as the applied voltage increases, representing that the device gives a capacitive response to the low applied AC voltage and the low temperature. And it was found that higher applied voltage gives a resistive response.

Real part of impedance was confirmed to decrease with increasing temperature. The first plateau shows complex impedance Z and the second plateau was a contact resistance Rs. And the gradual drop region was a complex component of capacitance and resistance. It was also confirmed that the magnitude of Z' of the first plateau is about 9 kΩ ~ 10 kΩ and the second plateau is about 20 Ω. Imaginary part of impedance was confirmed to decrease with increasing temperature too. The Core-Cole plot shows the dielectric relaxation at low frequency and complex impedance decreased depending on temperature with radius of R_p.

Acknowledgement

This work was supported by the Gachon University research fund of 2012. (GCU-2012-R037).

References

1. I.D. Parker, J. Appl. Phys. 75 [3] (1994) 1658.
2. C. Jonda and A.B.R. Mayer, Chem. Mater. 11 (1999) 2429.
3. S. Roy, S. Kundu, S.K. Roy, A.J. Pal, Materials Chem. and Phys. 77 (2002) 784.
4. D.H. Chung, T.W. Kim., TEEM 8 [3] (2007) 131.