OLED degradation mechanism study using impedance spectroscopy

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Abstract

To the best of our knowledge, for the first time, we applied impedance spectroscopy to analysis on OLED degradation mechanism by monitoring impedance change during constant voltage aging, and modeling OLED with lumped circuit elements. Change in each element value was used to explain charge accumulation and field redistribution in each organic layer.

1. Introduction

Impedance spectroscopy (IS) is a powerful tool to analyze electrical characteristics of black box system or materials by measuring frequency-dependent impedance[1], which has been widely used to analyze negative capacitance (NC) of low-mobility organic or inorganic thin-film devices[2]. NC has been typically explained by minority carrier flow, interface states, slow transient time of injected carriers, charge trapping or space charge. For OLEDs, IS has been used to explain operation of multi-layer device and corresponding electrical modeling with lumped circuit elements such as resistors, capacitors, and inductors (equivalence to the NC element)[3]. By changing device physical structure, change in each circuit element value was measured and related to hole injection behavior and thicknesses of emissive and hole transport layers. In fact, since charge accumulation and field redistribution in aged OLED can significantly affect carrier injection and transport through the device, carriers in the aged device will indirectly experience change in effective physical thickness of each layer and in time constant response at NC related interfaces. Therefore, we applied IS to study OLED degradation mechanism in this paper. Impedance change was monitored for a standard tris-(8-hydroxyquinoline) aluminum (Alq_3) based OLED during a constant voltage aging test. By extracting changes in resistors, capacitors, and NC elements, we related impedance changes to charge accumulation and field redistribution inside the OLED.

2. Experimental

In this study, we used a bottom emission fluorescent OLED, where we used indium-tin-oxide (ITO) as anode material, 4,4'-bis [N-(1-napthyl)-N-phenyl-amino] biphenyl (α-NPB) as hole transport layer (HTL) material, Alq_3 as electron transfer layer/light emissive layer (ETL/LEL) material, and bi-layer of lithium fluoride (LiF) / aluminum (Al) as cathode material, as shown in Fig. 1. Thicknesses of organic and cathode layers were 60, 70, and 0.5/100 nm, respectively. Green light mission was obtained from the fabricated OLEDs. ITO patterned glass substrates were cleaned according to a conventional cleaning process, treated with ultraviolet (UV) ozone for 15 minutes, and then annealed on a hot plate for 10 minutes. Organic layers and cathode layer were consequently deposited by a thermal evaporation method without breaking vacuum under a high vacuum condition (2.6~4.8 × 10^{-6} Torr). All the OLEDs were encapsulated by glass caps with desiccant. The light emitting area was 4 mm^2.

For OLED current-voltage (I-V) characteristics measurement, we used a semiconductor parameter analyzer, HP 4155C. I-V characteristics were measured and compared as shown in Fig. 2 before and after all bias stress and impedance measurement sequences. OLED impedance was measured by using a LCR meter, HP4284A, which has a bandwidth of 1MHz. The impedance was measured at 7 V DC with 400 mV rms AC oscillation signal every hour during the bias stress test, where a constant voltage (7 V) was applied from HP4284A as the stress bias signal. We used a constant voltage as the bias stress signal because the constant current bias stress was not possible for our Labview-based automatic stress and measurement setup. Total bias stress time was about 23 hours. All the measurements were performed at room temperature in air.

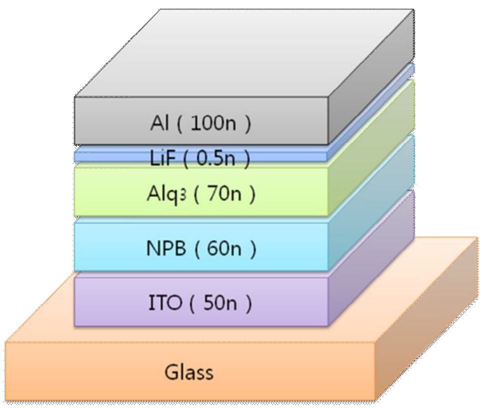

Figure 1. Structure of the fabricated OLED

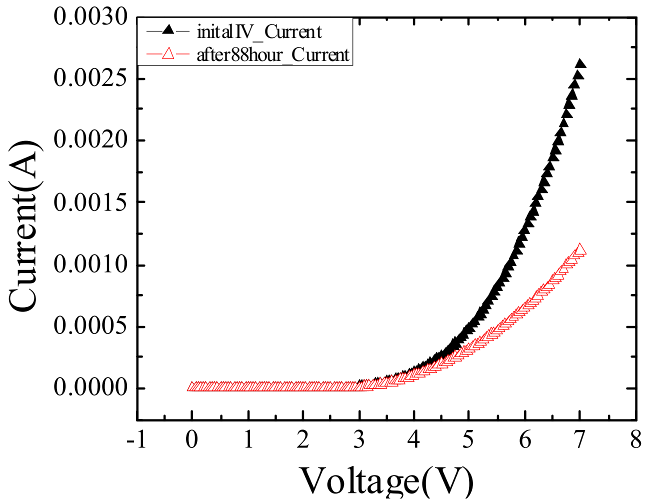

Figure 2. I-V characteristics of the fabricated OLEDs before and after the bias stress test for 23 hours

3. Result and Discussion

Figure 3 shows Cole-Cole plots of the measured OLED impedance and their changes with the stress time. Horizontal and vertical axes represent resistance and reactance, respectively. Positive reactance was observed at low frequency for both fresh and aged devices. This positive reactance is directly related to the NC of OLEDs, which was previously reported by other groups. Over the frequency ranges where the OLED has positive reactance, resistance component increases from about 500 ohm until the OLED reactance becomes negative. As the frequency further increases, the OLED resistance decreases while the reactance increases in its magnitude from zero up to certain frequency range and then decreases toward zero. We used the frequency range from 20 Hz to 1 MHz, which is a typical frequency range available from HP4284A. As the stress time increases, the contour of the Cole-Cole plots spreads outwardly as indicated with black arrows in the figure. If we have a good circuit model representing this Cole-Cole plot results, we can extract parameter values of each element in the model by using a conventional mathematical program tool. Although there have been many attempts to accurately model the Alq_3-based standard OLEDs[4,5,6], Pingree's equivalent circuit model best represents the NC behavior of the OLEDs, where Alq_3 layer was modeled as a parallel connection of a resistor (R_C) and a capacitor (C), α-NPB bulk layer was modeled as a parallel connection of a resistor (R_{L1}) and an inductor (L_1), and the interface between α-NPB and ITO anode as a series connection of a resistor (R_{L2}) and an inductor (L_2), as shown in Fig. 4[3,7]. ITO is represented as a resistor (R_S)

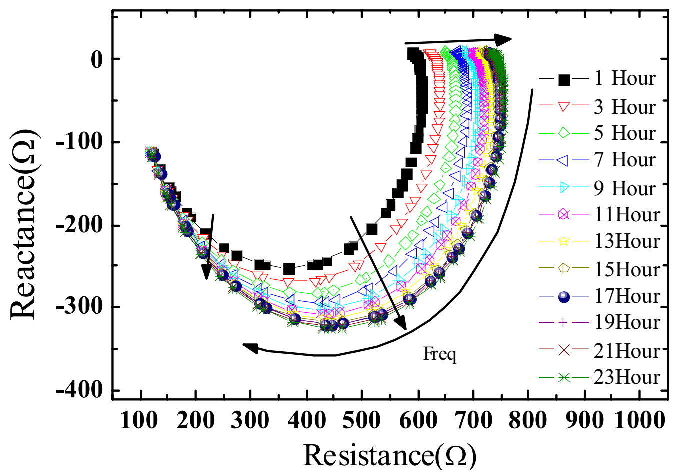

Figure 3. Cole-Cole plots of the measured OLED impedance and their changes with the stress time

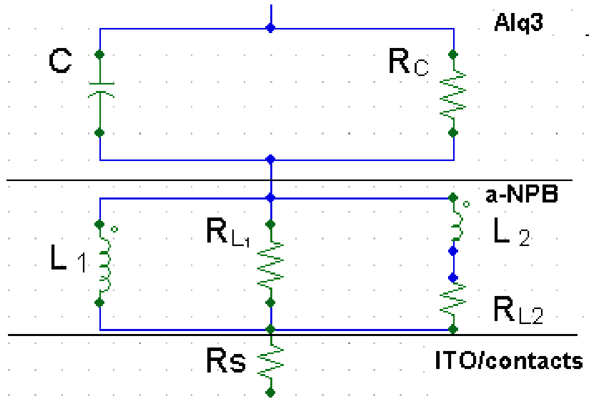

Figure 4. OLED equivalent circuit model used in this study

Total impedance of the OLED equivalent circuit model can be represented as the following equation.

$$Z_{total} = R_S + \left(R_C \parallel \frac{1}{j\omega C}\right) + \left(R_{L1} \parallel j\omega L_1 \parallel \left(R_{L2} + j\omega L_2\right)\right) \quad (1)$$

By comparing impedance behaviors of both equation (1) and measured data, we were able to successfully extract each element value. Figure 5 shows changes in each extracted element value with the stress time. While all the resistance and inductance increased with the stress time, capacitance decreases with the stress time. Although it is very difficult to accurately relate each parameter change with the degradation behavior and pose any physical meaning on the change, some of the parameter change can be explained as follows. Change of R_C can be considered as change in the bulk transport properties of the Alq_3 layer. R_{L1}, R_{L2} can be also explained in a similar manner. As the organic material and layer degrades, defect or trap sites can increase in the bulk layers, impeding charge carrier movement and thus, resulting in changes in resistance of the bulk layers. As mentioned before, since the NC is related to various physical phenomenon, we do not have a clear explanation for the changes in L_1 and L_2, which are under current intense investigation. However, both inductance values increase with the stress time and it is speculated that this phenomenon is related to the charge accumulation at the organic interface and corresponding electric field redistribution in the OLED as the device degrades.

Regarding change in capacitance value with the stress time, the extracted value decreased from 1.49 nF to 1.44 nF after the OLED degraded. Capacitance

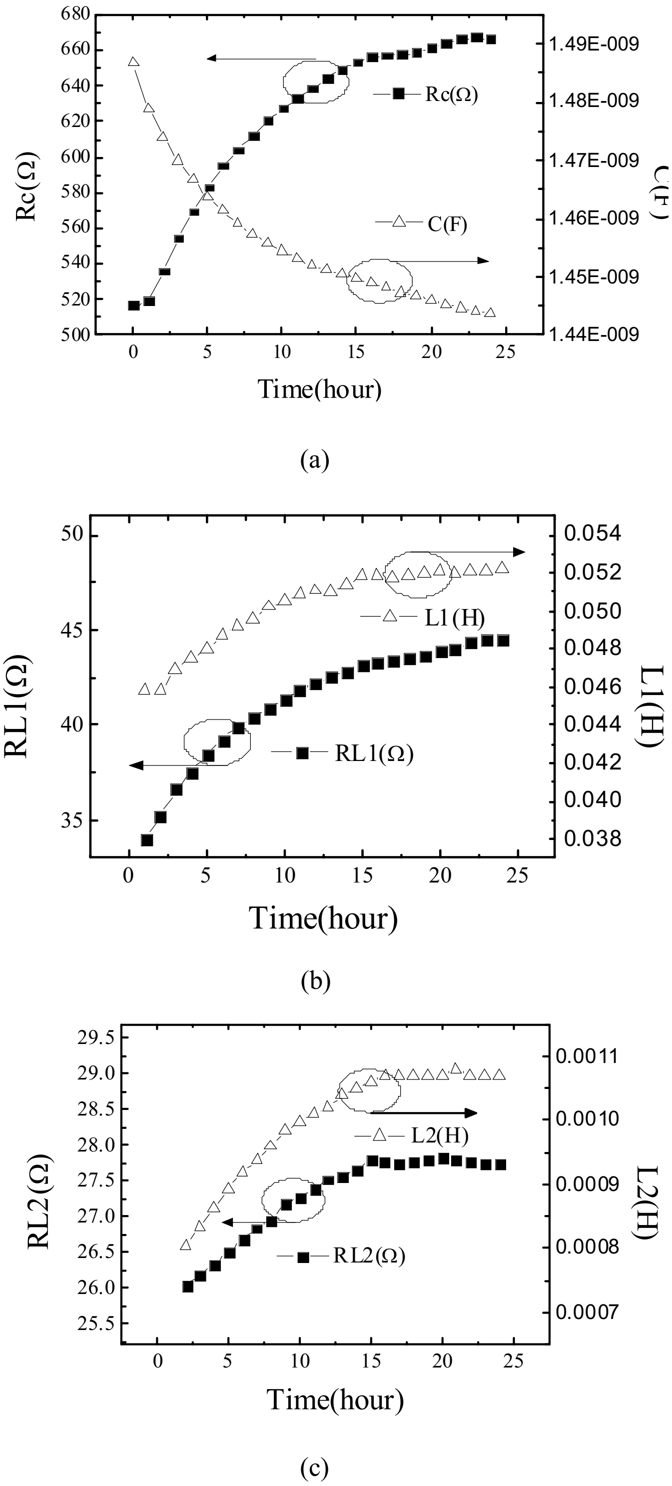

Figure 5. Changes in extracted parameter values with the stress time for (a) resistor and capacitor modeled for Alq3 layer, resistor and inductor modeled for (b) α-NPB bulk & (c) interface of α-NPB/Alq3. All the parameters were extracted from Cole-Cole plot of the measured data and equation

(1).
is closely related to dielectric constant, area, and thickness of the Alq_3 layer. It is assumed that there is negligible change in dielectric constant and area during the stress time period that we are considering. In fact, when we observed the aged device under the microscope, we did not observed reduction in the light-emitting area of our OLEDs. The thickness or effective distance for electron transport from cathode to the recombination zone possibly changed as the device degraded. Degradation induced recombination zone shift or change of the light-emitting zone depth change with degradation have been already reported for mixed host[8] and conventional Alq/diamine (TPD)[9] based OLEDs, respectively. In the mixed host OLED, it was reported that the recombination zone was formed in the light-emitting layer 10 nm away from the interface of light-emitting layer and hole transport layer, and then the recombination zone shifted to the interface and became broaden as the device degraded. For the conventional Alq based OLEDs, it was reported that the recombination zone was distributed in the Alq layer up to about 20 nm away from the Alq/TPD interface and shrank toward the interface as the device degraded. Although we do not have experimental results for this type of recombination zone variation with the stress time at this stage, we believe that in our device, there should be also similar shift or shrinkage of the recombination zone as the device degrades, which can effectively change the distance for electron transport from cathode to the recombination zone, resulting in increase and decrease of R_C and C, respectively.

4. Summary

We used impedance spectroscopy method for the first time to investigate OLED degradation behavior by using lumped element circuit model and extracting each parameter values. Changes in each parameter with the stress time were explained by using charge accumulation and field redistribution at the organic/organic interface, which is one of the major OLED degradation mechanisms. Although, in circuit model, it is assumed that each element separately represents organic bulk layer or interface and there is still more work to utilize the impedance changes in explaining complicated device physics, we believe that the impedance spectroscopy is a very useful tool to analyze the OLED operation and degradation behaviors.

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