

Effect of HfO_x treatment on ITO surface of organic light emitting diodes using Impedance spectroscopy analysis

Jaehyun Cho¹, Hyungjun Park¹, Kyumin Han¹, Sun Young Sohn², Dong Geun Jung² and Junsin Yi*

¹School of information and communication engineering, Sungkyunkwan University, Suwon, 440-746, Korea

²Department of Physics, Institute of Basic Science, and Brain Korea 21 Physics Research Division, Sungkyunkwan University, Suwon 440-746, Korea

Keywords : Organic light emitting diodes, HfO_x , Impedance spectroscopy analysis, Equivalent circuit, Barrier height

Abstract

In this work, we used impedance spectroscopy analysis to determine the effect of the HfO_x treatment on the surface of ITO and to model the equivalent circuit for OLEDs. Devices with an ITO/Organic material/Al structure can be modeled as resistances and capacitances arranged in parallel or in series. The number of elements depends on the composition of the structure, essentially the number of layers, and the contacts.

1. Introduction

In organic light emitting diodes (OLEDs), both the electrons and holes need to be injected efficiently to obtain the best device performance. This means that a small injection barrier height at the indium-tin oxide (ITO)/organic interface is required. The insertion of an insulating layer between the ITO and organic layers leads to a significant improvement in the charge injection and electroluminescence output. In this study, the surface of the ITO anode in OLEDs was treated with an HfO_x deposition process using an atomic layer chemical vapor deposition system (ALCVD). The OLEDs fabricated on the HfO_x treated ITO anode showed a lower impedance and higher conductance and capacitance. The change in capacitance, conductance and impedance was attributed to the enhanced carrier tunneling and to the change in the work function of ITO.

2. Experimental

ITO (150nm)-coated glass with a sheet resistance of $20 \Omega/\square$ was used as the anode for the fabrication of the OLED. In this process, the ITO glass was cleaned sequentially in ultrasonic baths consisting of trichloroethylene, acetone, and methanol, then sonicated in deionized water and finally blown dry with N_2 gas. To modify the surface of the ITO samples, they were treated using atomic layer deposition with tetrakis(ethylmethylamino) hafnium (TEMAH). For the ALCVD treatment, each cycle was composed of four steps as follows. First, a mixture of TEMAH and Ar carrier gas (300 sccm) was introduced into

the vacuum chamber for 10 s. Then, the chamber was purged by flowing Ar (150 sccm) for 5 s. For the formation of the HfO_x layer, oxygen gas (300 sccm) was allowed to flow for 5 s, and the resulting by-products or remnant gases were completely removed by the subsequent purging step using Ar (150 sccm) gas for 5 s. The ITO substrates were then treated in an ALCVD system for 5 cycles at room temperature (RT). After the surface treatments, *N,N'*-diphenyl-*N,N'*-bis(3-methylphenyl)-1,1'-diphenyl-4,4'-diamine (TPD, 32 nm), tris-(8-hydroxyquinoline) aluminum (Alq_3 , 48 nm), lithium fluoride (LiF, 0.5 nm), and Al (90 nm) were sequentially deposited as the hole transport layer (HTL), the emitting material layer (EML), the cathode interfacial layer, and the cathode, respectively. The device configuration of ITO/ HfO_x / TPD/ Alq_3 / LiF/Al is shown in Fig. 1. For the electrical measurements, the capacitance, conductance and impedance measurements were carried out using an LF 4192A impedance analyzer.

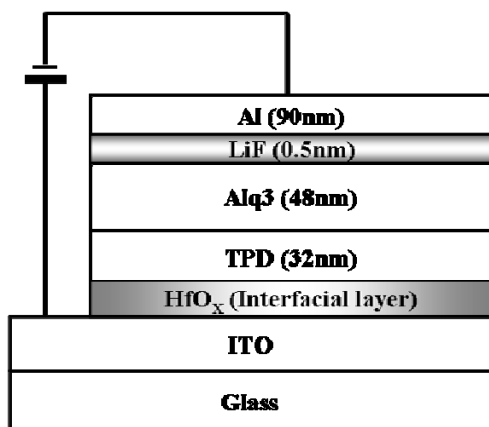


Fig. 1. The structure of the OLEDs with the HfO_x treatment on the surface of the ITO anode

3. Results and discussion

For the electrical measurements, the capacitance, conductance and impedance measurements were carried out using an LF 4192A impedance analyzer. The amplitude of the test signal was 50 mV. The measurement frequency was in the range of 200 Hz to 10 MHz. Figures 2 (a) and (b) show the capacitance versus frequency (C-F) and the conductance versus frequency (G-F) characteristics, respectively, for the devices with the HfO_x treated and non-treated ITO anodes. The capacitance of the HfO_x treated devices is slightly better than that of the non-treated ones. This is related to the enhancement of the carrier injection, because the capacitance depends on the amount of charge injection into the organic layer from the electrodes[3]. The conductance of the HfO_x treatment sample was slightly increased compared to that of the OLEDs without any treatment. In the high frequency region, the values of the conductance are almost the same, whereas, in the low frequency region, there is an increment in the conductance brought about by the HfO_x treatment of the ITO surface. This suggests that the hole injection barrier is reduced due to the alignment of the work function of ITO.

For the purpose of building an equivalent circuit model of the OLEDs, the impedance of the device was measured in the range of 200Hz to 10MHz at zero bias voltage. The values were analyzed using the following complex impedance equation [4]. Figures 3(a) and (b) show the frequency-dependent real and imaginary parts of the impedance of the devices with the HfO_x treated and non-treated ITO anodes, respectively. The impedance of the OLEDs simulated with the equivalent circuit model of Fig. 3(c) show

substantial correspondence, as compared with the measured results shown in Fig. 1, where the measured and simulated impedances are depicted as symbols and lines, respectively. The equivalent circuit of these devices consists of an RC network in series with a contact resistance (R_s). As shown in Figures 3(a) and (b), this real part of the impedance for the devices with the HfO_x treated ITO anode is lower than that in the devices with the non-treated ITO. The real part of the impedance of the devices with the HfO_x treated ITO is significantly lower in the high frequency regions. We developed an equivalent electrical circuit for a two-layered structure, in which TPD is used for the hole transport layer and Alq_3 for the electron transport and light emitting layer, which consists of two parallel RC circuits connected in series, as shown in Figure 3(c). The simulated results are in very good accordance with the experimental results.

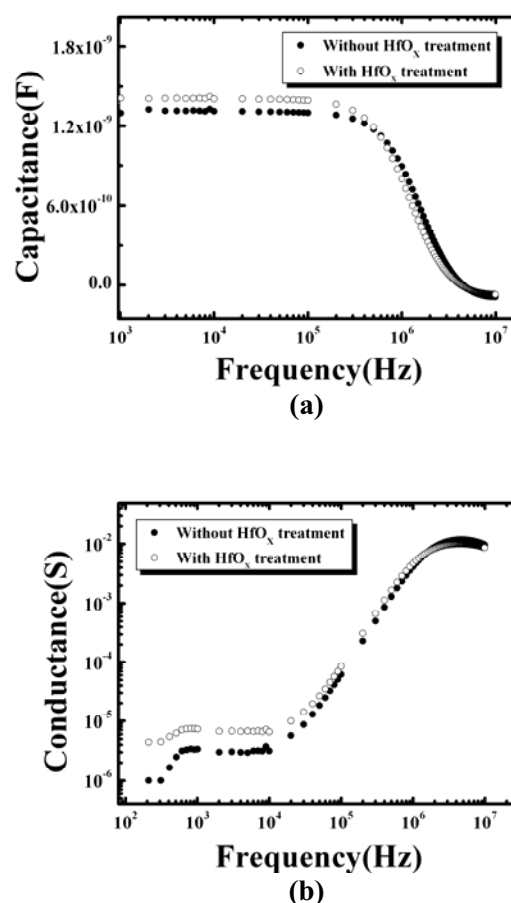


Fig. 2. Properties of OLEDs fabricated on the ITO anode subjected to HfO_x treatment (a) Capacitance variation with frequency and (b) Conductance variation with frequency.

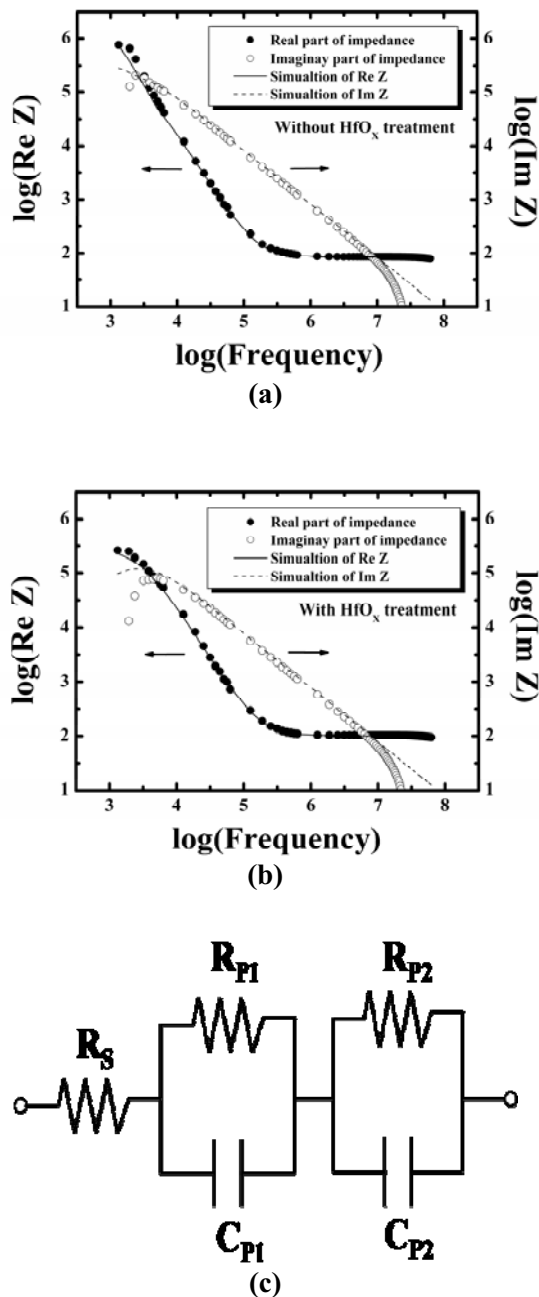


Fig. 3. The frequency-dependant real and imaginary parts of the impedance of the OLEDs (a) without and (b) with HfO_x treatment for 5 cycles at room temperature. The symbols are the measured data and the solid lines are the fitting data using the equivalent circuit by the model of (c).

4. Summary

In this study, the effects of HfO_x treatment of the ITO anode on the electrical characteristics of OLEDs

were investigated. OLEDs were fabricated with TEMAH treated ITO substrates using the ALCVD process. Through the insertion of an HfO_x film as an interfacial layer between the ITO anode and HTL brought about by treating the ITO anode with HfO_x for 5 cycles at room temperature, the performance of the OLEDs was improved. The device with the HfO_x interfacial layer at the ITO/TPD interface can be modeled as a simple combination of an RC network in series with a resistance and capacitance whose values are decreased and increased, respectively, in comparison to those of the device without the anode interfacial layer. This enhancement can be attributed to the improvement of the hole injection and alignment of the work function of ITO. The simulated results of this equivalent circuit model are in very good agreement with the experimental results and this model can be used for the modeling of OLEDs.

5. References

1. Y. S. Lee, J. H. Park, J. S. Choi, and J. I. Han, *Jpn. J. Appl. Phys.* **42**, 2715 (2003).
2. S. H. Kim, K. H. Choi, H. M. Lee, D. H. Hwang, L. M. Do, H. Y. Chu, and T. Zyung, *J. Appl. Phys.* **87**, 882 (2000).
3. V. Shrotriya and Y. Yang, *J. Appl. Phys.* **97**, 54504 (2005).
4. J. Ross Macdonald, "Impedance Spectroscopy-Emphasizing Solid Materials and Systems", (John wiley & Sons, New York, 1987)