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Influence of Copper Phthalocyanine (CuPc) Thin Layer on Capacitance-Voltage Characterization of a Device Consisting of ITO/CuPc/PVK/Rhodamine B Dye Layers

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Abstract: In this paper, the influence of copper-phthalocyanine (CuPc) hole-injection layer on the capacitance-voltage (C-V) characteristic performance of ITO/CuPc/PVK/Rhodamine B/Pb devices was investigated. The thickness of the CuPc layer was varied from 5 nm to 50 nm. The thin films were deposited on the PVK layer by thermal vacuum deposition at 1.0×10^{-5} torr using MiniLab 080 system. The thicknesses were measured using FILMETRICS F20-UVX thin-film analyzer. It was found that as the thickness of CuPc layer decreases the transition voltage (V_T) and built-in voltage (V_{bi}) decrease except for a thickness 5 nm, which shows negative differential resistance (NDR).

Keywords: OLED, Copper-Phthalocyanine, J-V Curves, C-V curves.

1 Introduction

Organic Light-Emitting Diodes (OLEDs) have been recognized as a promising technology for large-area, full-color and flexible displays. A lot of effort has been made to improve the device structures and to understand their operating mechanisms [1-14]. The light emitted by an OLED is generated through the process of electroluminescence. This phenomenon arises from the recombination of electron-hole that occurs under applying a voltage to a semiconductor material sandwiched between two electrodes. When a voltage is applied to an OLED, electrons are injected from the cathode into electron-transport layer, at the same time holes are injected from the anode into the hole-transport layer. Then charge carriers transport through the layers until they recombine in the emission layer and emit light.

The basic current structure of OLED has a high work function conducting transparent anode, one or more organic layer and low work function cathode. Most OLEDs use ITO as anode because it has high work function and high transparency (90%) to visible light. Another important parameter is its work function (Φ_0) or Fermi energy level (E_F) relative to the organic material, a high Φ_0 is needed for the anode to have more efficient OLED. On the other hand, a low work function of cathode is required to allow a lot of electrons to inject into the organic layer for efficient OLEDs.

The performance of OLED devices was shown to be improved by introducing a thin layer of phthalocyanine (CuPc) between the anode and the hole-transport layer [1].

Emission properties of OLEDs were investigated with the use of a hole-injection layer of CuPc [2].

In 2006, Po-Ching Kao et al. studied the influence of the metal-phthalocyanine (MPc) hole-injection layer on the electroluminescence performance of ITO / MPc / Alq₃ / Al devices [5]. They found that the turn-on voltage (V_t) is lowered by inserting MPc layers and remains virtually the same as the MPc layer thickness is adjusted in the range of 5-15nm. In addition, the turn-on voltage decreases significantly with the increase of the HOMO levels of the MPc films. It was found that the bias dependent capacitance of organic hetero-layer light emitting diodes based on N, N'-diphenyl-N, N'-bis (1-naphthyl)-1-biphenyl-4, 4 diamine (NPB) and tris (8-hydroxyquinoline) aluminium (Alq₃) shows below the built-in voltage, a step-like change from a value corresponding to the total organic layer thickness to a higher value given by the Alq₃ layer thickness [6]. V. Shrotriya and Y. Yang studied the influence of several metals, such as calcium (Ca), aluminum (Al) and gold (Cu), as the cathode into the charge injection on the C-V characteristics [9]. In 2009, S. Nowy et al. employed the impedance spectroscopy to investigate the charge carrier injection properties of different anode and anode treatments in bottom-emitting organic light-emitting diodes (OLEDs) [10].

In this paper, we report the (C-V) characteristics of OLEDs fabricated by adding CuPc layer into the device consisting of ITO/ PVK/Rhodamine B dye/Pb, and relate the behavior of C-V characteristics to the onset of majority and minority charges injection into the active organic layer from the

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electrodes. Compared to the C-V curves, the current density-voltage (J-V) curves give very limited information about the physical processes in OLEDs.

2 Experiments

ITO-glass substrates with dimension $2.5\text{cm} \times 2.5\text{cm}$ were used as anode layer. The substrates were chemically cleaned using acetone, isopropanol and distilled water in an ultrasonic cleaner. After the chemical cleaning, the ITO glass substrates were placed in an oven at 60°C for 24 hours in order to remove the residual water. In this work, two types of devices were fabricated. The first is a double layer with the structure ITO / PVK / Rhodamine B dye / Pb as shown in Fig. 1.

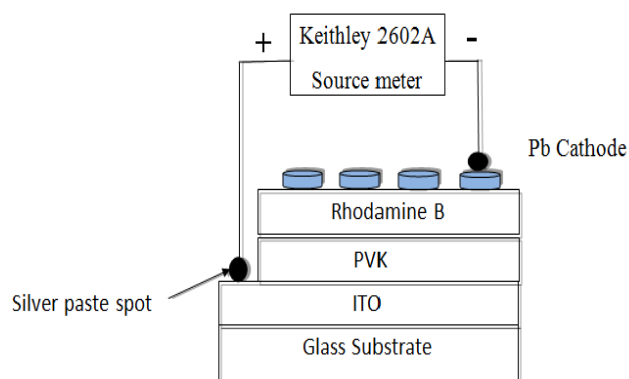


Figure 1: Schematic drawing of a double-layer ITO/PVK/Rhodamine B/Pb OLED.

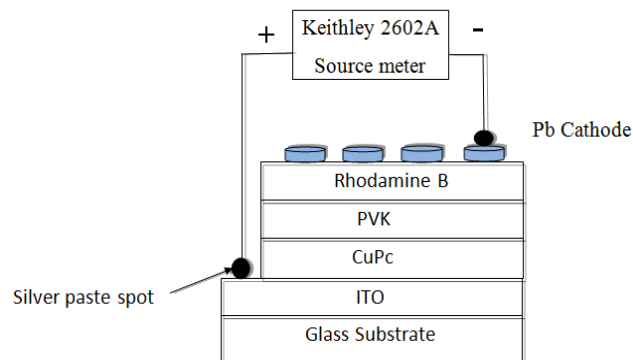


Figure 2: Schematic drawing of a multilayer ITO/CuPc/PVK/Rhodamine B/Pb OLED.

15 mg of PVK powder was dissolved in mixed solvents consisting of 5ml tetrahydrofuran (THF) and 1ml toluene. A homogenous solution was obtained and deposited on the ITO substrate. The substrate was inclined at an angle of 45° for several minutes. The solution spreads on the surface of the substrate due to gravity to form a thin film of thickness of about 45nm. This process is sensitive to the variation in temperature due to different evaporation rates of solvent [13]. The thickness of the PVK layer was measured using

FILMETRICS F20-UVX thin-film analyzer. A 30nm Rhodamine B dye layer is then deposited on the PVK layer by thermal vacuum deposition at 1.0×10^{-5} torr using MiniLab 080 system made by Moorfield Associates, UK. Finally, 35nm Pb was deposited as a cathode electrode on Rhodamine B dye layer. The negative terminal of the applied voltage was connected to the Pb electrode through a drop of Hg whereas the positive terminal was connected to ITO substrate by silver paste. The second device contains a multilayer with the structure ITO / CuPc / PVK / Rhodamine B dye / Pb as shown in Fig. 2. After cleaning the ITO surface, CuPc layer was deposited by thermal vacuum evaporation at 1.0×10^{-5} torr. PVK and Rhodamine B layers were deposited on the CuPc layer as described above in the first device.

The voltage-current signals were applied and recorded through a Keithley 2602A source meter that was used to control, measure the voltage across the sample and the current flowing through it. The C-V curves were measured and recorded by GW 821 LCR meter for the voltage range from 0V up to 30V. Electrical measurements were carried out at room temperature and normal ambient conditions. The relative light intensity emitted from the samples was measured using a photodiode. The output of the photodiode was amplified and collected by the Keithley 2602A source meter.

3 Results and Discussion

Figures 3 and 4 represent the energy level diagram of the fabricated OLED devices. As can be seen from the band models, adding the CuPc layer between ITO and PVK layer reduces the hole barrier for holes injected from the anode into organic layer. The injected holes become dominant charge carriers and make the device increasingly single carrier "hole-only". This means that the current density in the device is controlled almost by the holes [9]. The barrier to hole injection at the anode is obtained by the equation $\Delta E_h = IP - \Phi_A$ where Φ_A is work function of anode and IP is the ionization potential or HOMO level relative to the anode. We find that $\Delta E_h = -1\text{eV}$ for the first device with the structure ITO / PVK / Rhodamine B dye / Pb (Fig. 3). As shown in Fig. 4, the hole injection barrier (ΔE_h) of the device in the presence of the CuPc layer is about -0.4eV . There is a reduction of about 0.6eV due to the presence of CuPc layer.

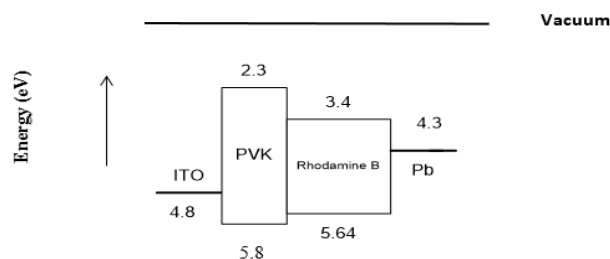


Figure 3: Energy level diagram for the device consisting of ITO/PVK/Rhodamine B dye/Pb.

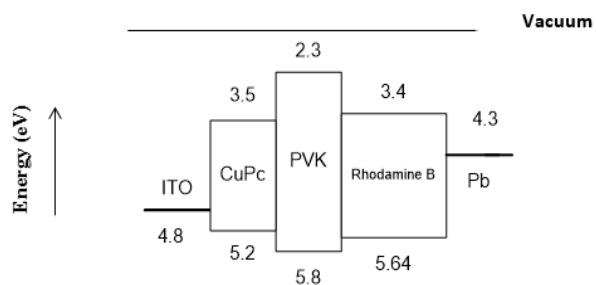


Figure 4: Energy level diagram for the device consisting of ITO/ CuPc / PVK / Rhodamine B dye/Pb.

Figure 5 shows a typical C-V curve for an OLED device. The voltage V_1 is the bias voltage at which the majority charge injection into the organic layer starts in forward bias and sometime called transition voltage (V_T) [9]. Below this voltage, the organic layer is neutral with a very small amount of charge trapped in the defect sites near the interface. The organic layer can be thought of as composed of small charge regions near the interface and large neutral region elsewhere. Above V_1 , a sharp capacitance increase is observed corresponding to the majority charge injection. The voltage V_2 corresponds to the device turn-on voltage, at which the light emission from the organic layer begins and marks a sharp decline in the value of the measured capacitance of the device [9]. The voltage V_2 or sometime called built-in voltage (V_{bi}) is the bias voltage at which the minority charge carriers starts injection into the organic layer.

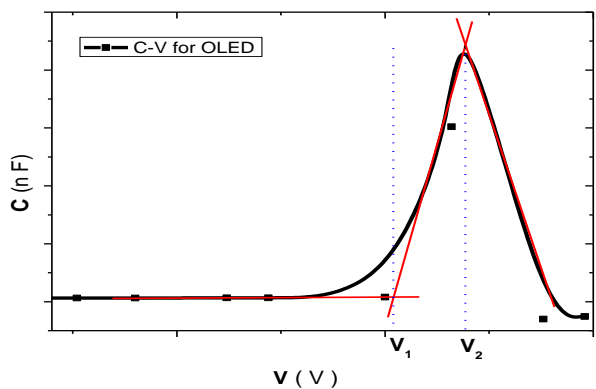


Figure 5: A typical C-V curve for an OLED device under forward applied bias.

Figure 6 shows the capacitance-voltage curve for the control device with the structure of ITO/45nm PVK/30nm Rhodamine B dye/35nm Pb. It is clear that the C-V characteristic curve shows minor and major peaks which refers to the injection of charges into the device.

At the minor peak, the capacitance increases at transition voltage V_{T1} which approximately at 7.0 V due to the injection of holes, where at voltage of about 7.7 V which is called the built-in voltage V_{bi1} some electrons can be injected. The capacitance then decreases until the transition voltage of the major peak. For the major peak, holes are

injected into the PVK layer at transition voltage $V_{T2} = 8.6$ V. At $V_{bi2} = 9.7$ V, electrons are injected from the cathode into the device. As a result, electrons and holes recombine to generate the light and the charge is annihilated.

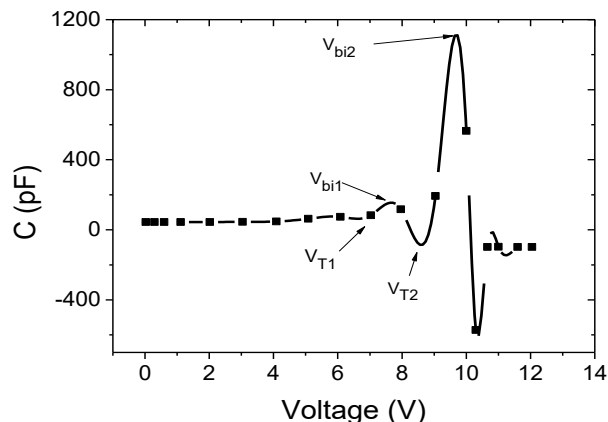


Figure 6: Capacitance- voltage characteristic curve of the standard device consisting of ITO/ 45nm PVK/ 30nm Rhodamine B/ 35nm Pb.

Table 1: The transition and built-in voltages for different CuPc layer thickness

Thickness of CuPc (nm)	0	5	10	30	50
V_{T1}	7.0	10.4	1.3	5.5	11.5
V_{T2}	8.6	12.2	4.2	12.0	14.5
V_{bi1}	7.7	11.0	1.9	7.5	13.6
V_{bi2}	9.7	13.0	6.8	13.4	15.4

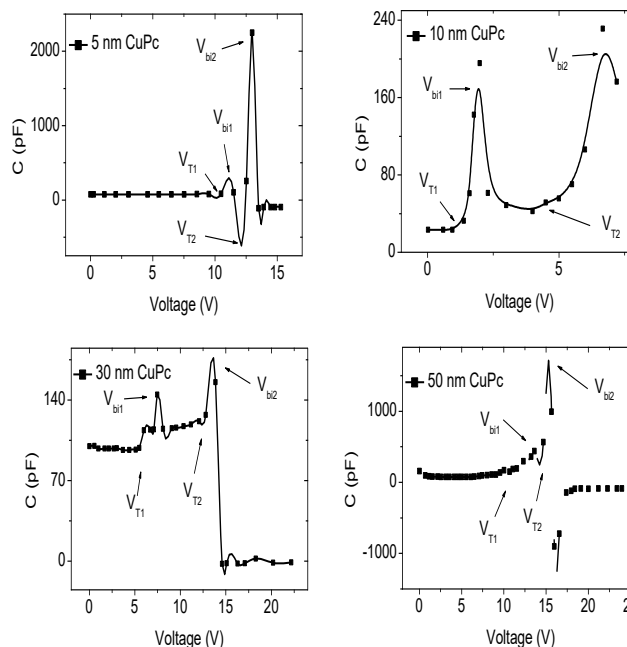


Figure 7: Capacitance- voltage characteristic curves of the device consisting of ITO/(5, 10, 30, and 50 nm) CuPc/ 45nm PVK/ 30nm Rhodamine B / 35nm Pb.

Figure 7 illustrates the C-V curves for devices with the structure of ITO/different thicknesses of CuPc/45nm PVK/30nm Rhodamine B dye/35nm Pb.

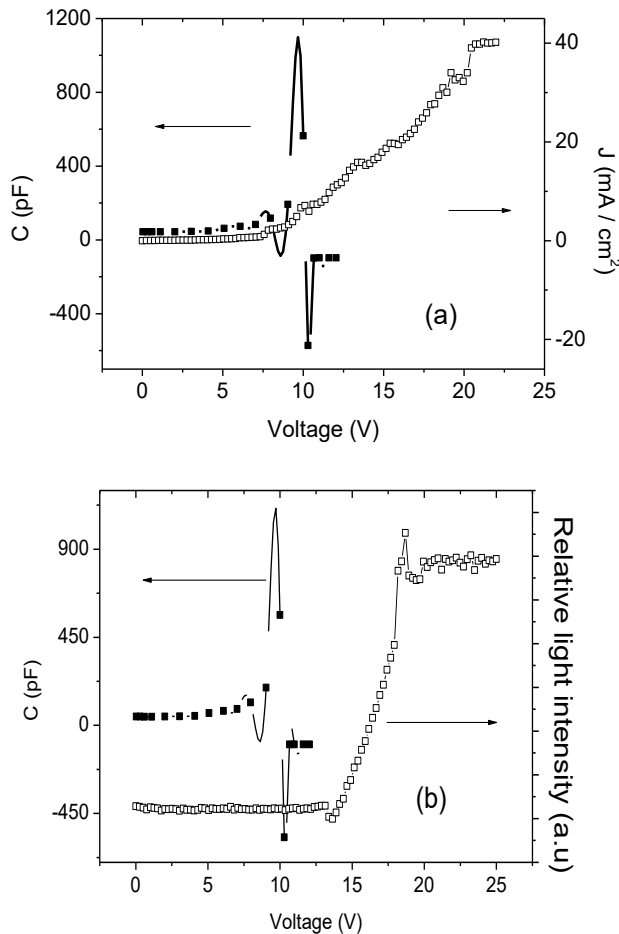


Figure 8: Current density-capacitance curve (a) and relative light intensity-capacitance curve (b) as a function of the applied forward bias for ITO/~44.88nm PVK/30nm Rhodamine B/35nm Pb device.

For a thickness of 5nm of the CuPc layer the transition voltages V_{T1} and V_{T2} at which holes are injected to the CuPc layer are 10.4 V, 12.2 V, respectively. On the other hand, the built-in voltages V_{bi1} , V_{bi2} at which electrons are injected into the device are 11.0 V and 13.0 V, respectively. For a thickness of 10 nm of CuPc the V_{T1} , and V_{T2} are 1.3 V and 4.2 V, respectively, the V_{bi1} , and V_{bi2} are 1.9 V and 6.8 V respectively. In the case of 30nm CuPc, the voltage at which holes can be injected (V_{T1} , and V_{T2}) are at 5.5 V, 12.0 V, respectively. On the other hand, at $V_{bi1}= 7.5$ V and $V_{bi2} = 13.4$ V the electrons can be injected into the device. Finally, for a thickness of 50nm of the CuPc layer the transition voltages V_{T1} and V_{T2} are 11.5V, 14.5 V, respectively, and the built-in voltages V_{bi1} , V_{bi2} are 13.6 V and 15.4 V, respectively. As the thickness of CuPc layer decreases the V_T and V_{bi} decrease as shown in Table 1, except for a thickness 5 nm, which shows negative differential resistance (NDR).

Figure 8 show the current density-capacitance curve (a) and relative light intensity-capacitance curve (b) as a function of the applied forward bias for the control device without the CuPc layer. At a positive bias of approximately $V_{bi1}= 7.7$ V, the current density drastically increases. This increase in current density is due to the injection of holes and electrons into the device. On the other hand, the light emission is observed at the same value of the applied potential at which the capacitance starts decreasing due to the recombination of holes and electrons, annihilating charges. Sometime the experimental quantities for V_{on} voltage can be changed to the V_{th} and V_{bi} voltages by some tenths of a volt as clearly seen in Fig. 8 (a) and (b) [10].

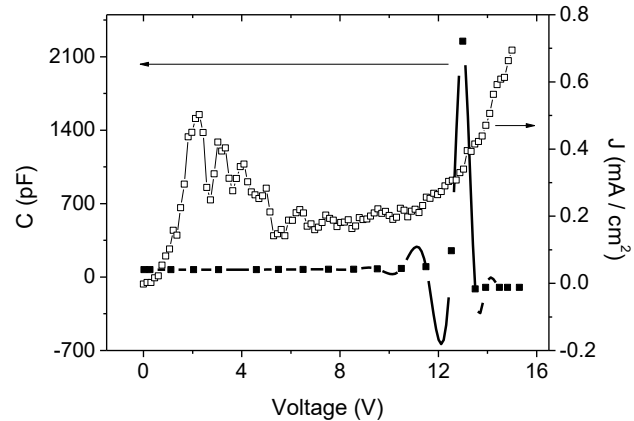


Figure 9: Current density-capacitance curves as a function of the applied forward bias for the device consisting of ITO/5nm CuPc/~44.88nm PVK/30nm Rhodamine B dye/35nm Pb.

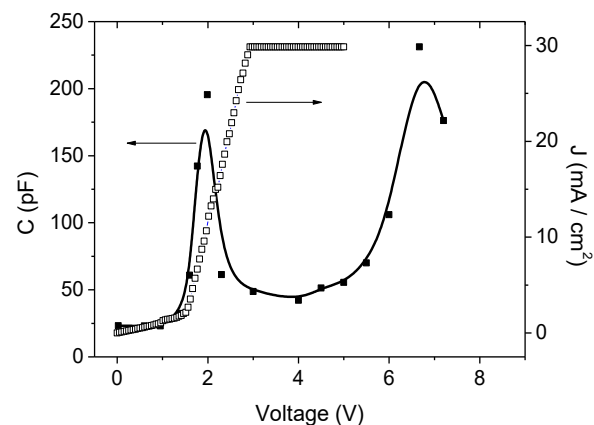


Figure 10: Current density-capacitance curves as a function of the applied forward bias for the device consisting of ITO/10nm CuPc/~44.88nm PVK/30nm Rhodamine B dye/35nm Pb.

Figures 9 through 12 show the current density-capacitance curve as a function of the applied forward bias for the devices having the structure ITO/(5nm, 10nm, 30nm, and 50nm) CuPc/ PVK/ Rhodamine B/Pb, respectively. As can be seen in the figures, when the current density begins to increase in

J-V curves, the capacitance also increases in C-V curves at the same voltage approximately.

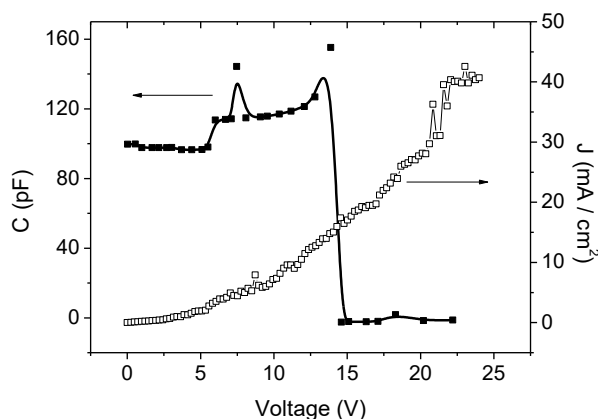


Figure 11: Current density-capacitance curves as a function of the applied forward bias for the device consisting of ITO/30nm CuPc/ ~44.88nm PVK/ 30nm Rhodamine B dye/ 35nm Pb.

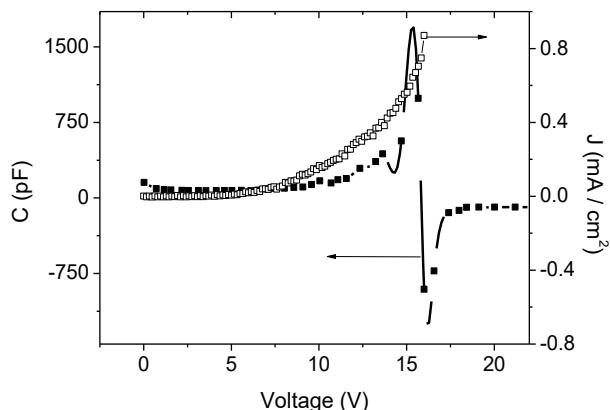


Figure 12: Current density-capacitance curves as a function of the applied forward bias for the device consisting of ITO/50nm CuPc/ ~44.88nm PVK/ 30nm Rhodamine B/ 35nm Pb.

4 Conclusion

In this paper, we have used impedance spectroscopy to study charge injection in OLED devices consisting of ITO/CuPc/PVK/Rhodamine B dye/Pb, with different layer thicknesses of CuPc. The effect of the CuPc layer on the transition voltage V_T and built-in voltage V_{bi} is as follows: when the thickness of the CuPc layer decreases, V_T and V_{bi} were observed to decrease expect at a thickness of 5 nm due to the negative differential resistance (NDR). We have shown that capacitance-voltage analysis is a sensitive tool to monitor the charge injection more than current density-voltage measurements.

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