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Investigation of the effect of two dyes on organic light emitting diodes electroluminescence

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Abstract: In this article, two organic light emitting diodes OLEDs have been fabricated and studied. Poly(9-vinylcarbazole) PVK doped with Rhodamine 6G and Carbocyanine dyes at weight concentration of 2% has been used as a hole transporting luminescent layer. Each film was sandwiched between indium tin oxide (ITO) and aluminum (Al) layers. The measurements include I-V characteristics and variation of relative light intensity with the driving voltage and with the current. It is found that the threshold voltage (V_T) of the samples is smaller than that obtained in such PVK-based devices, when the PVK film is obtained by spin coating. The thickness of doped layer as well as the optical parameters has been determined using variable angle spectroscopic ellipsometer.

Keywords: *Topological Organic light emitting diodes, PVK, Electroluminescence, ellipsometry.*

1. Introduction

Thin organic films have many attractive features and are being widely investigated by researchers for use in electronic devices. The main advantage of organic materials over inorganic semiconductors is that they can be deposited by evaporation, spin-coating, screen printing, and casting. These deposition methods are simpler and cheaper than most of the deposition techniques used in inorganic semiconductors.

Organic light emitting diodes (OLEDs) have been studied intensively since C. Tang and S. VanSlyka [1] reported their double layer organic light emitting device. In 1990, Burroughes *et al.* [2] made another important discovery when they detected electroluminescence (EL) from devices based on luminescent conjugated polymers.

An OLED is a display device that sandwiches single or multiple layers of organic thin films between two electrodes, one of the electrodes is a transparent indium tin oxide (ITO) anode, and the other is a metallic cathode. When a DC voltage is applied to an OLED, electrons and holes are injected from the cathode and the anode, respectively. The injected charge carriers may excite the luminescent material forming excitons which in turn give photons due to the hole-electron recombination [3-9]. The primary aspects of an OLED device performance are its perceived brightness or emissive ability, color tuning, durability, and control in operation.

Poly(9-vinylcarbazole) (PVK) is a well known polymer for its photoconducting properties [10-12]. It, also, exhibits photoluminescent properties in the UV region [2,4]. Djobo *et al.* [13] have studied single layer vacuum deposited PVK devices and reported a quite faint EL. They concluded that excitons are not confined within the PVK layer and that the use of an electron transport layer induces a red shift of the emitted light. Which is true since pure PVK is well known for transporting holes and blocking electrons.

In this article, we report EL from devices composed of a single PVK layer doped with Rhodamine 6G and Carbocyanine dyes at 2% weight concentration. The structures of PVK, Rhodamine 6G dye and Carbocyanine dye are shown in Fig. 1.

2. Experimental

ITO coated glass substrates purchased from delta technologies, USA with sheet resistance of 8–12 Ω/□ were cut carefully into pieces of 2.5 cm × 2.5 cm. Each sample was arranged in an 4 × 4 array of devices. The active area of each device was 2.2 mm × 2.2 mm. The ITO plates were cleaned thoroughly by wiping each electrode surface by a cotton pad wetted with acetone. Then, an electric tape is stuck at the conductive side of the ITO coated glass substrate. The ITO electrodes were then etched to form the desired pattern by immersing them in Aqua-Regia acid (solution of 20% HCl and 5% HNO_3) for 15 minutes at 60°C. A digital multimeter (Fluke77) was used to measure the resistance of the etched regions of the ITO substrates. Very high resistance must be recorded if the etching procedure is successful in removing the ITO layer from the etched regions. The electrodes were cleaned using Xyelen, then rinsed in deionized water to remove away the leftover acids on their surfaces. Upon completing the etching process, the electrodes were cleaned using DC argon plasma for 8 minutes. Then, the electrodes were left inside an oven at 80°C over night.

An amount of 30 mg of PVK (Sigma-Aldrich, USA) was dissolved in 15 ml of chloroform. An amount of 2% by weight of the dopant (Rhodamine 6G and Carbocyanine dyes) was added to the solution. The solution was then stirred thoroughly and filtered. Few drops of the blend solution were spun at 1750 rpm for one minute to form thin film on the ITO.

The samples were kept under vacuum at 80°C for 24 hours to remove residual solvents. Al cathodes were then deposited thermally under vacuum condition (10^{-5} torr) at a low deposition rate on the top of the structure through a five fingers shadow mask. Finally, wires were connected to the electrodes using silver glue. The structure of the device is shown in Fig.2.

A schematic diagram of the structure of the experimental setup is depicted in Fig.3. The relative light intensity was measured using a PMT (photomultiplier tube). The data were recorded and saved as an ASCII file, then, analyzed and plotted. Electrical measurements were conducted at room temperature and in nitrogen chamber in order to prevent oxidation and to increase the device life time. The driving voltages and currents were obtained and measured using a MetraByte's Das-20 data acquisition card interfaced with a personal computer. The output of the PMT was amplified then collected by the same data acquisition card.

The film thickness of the device (48 nm) as well as the optical properties PVK were measured using a homemade variable angle spectroscopic ellipsometer (VASE).

3. Results and Discussion

Ellipsometry is a sensitive optical technique for determining properties of surfaces and thin films. In this work we use a homemade variable angle spectroscopic ellipsometer (VASE) to determine the relative phase angle between p- and s-polarized lights, Δ, and the relative amplitude ratio, ψ, as well as the refractive index, n, and the thickness of the PVK film deposited on Si wafer. The film thickness was found to be 48 nm. Figure 4 depicts the variations of Δ and ψ with wavelength at an angle of incidence 75°. Figure 5 illustrates the variation of the refractive index of PVK film on Si wafer with wavelength (visible spectrum) at an angle of incidence 75°.

PVK is a well known hole transporting polymer that exhibits essentially trap-free transport for holes [3]. Accordingly, the injected holes can reach the Al electrode without recombination, unless they encounter electrons injected from the Al electrode.

As mentioned in the experimental work section, two single layer devices (S_1 and S_2) have been fabricated. S_1 device has the structure ITO/PVK:Rhodamine 6G/Al while S_2 device has the structure ITO/PVK:carbocyanine/Al. Figure 6 illustrates the I-V characteristics for S_1 and S_2. It is clear from the figure that the current increases exponentially with the applied voltage.

The threshold voltage, V_T, is an important parameter characterizing the operation of an OLED. In general it is the voltage at which an OLED starts to emit light. The current increases sharply when the applied voltage exceeds V_T. The threshold voltages were estimated by linearly extrapolating the I-V curves back to the voltage axis [7,8]. As can be seen from Fig. 6, V_T of S_1 (~ 4.3 volt) is less than that of S_2 (~ 7.9 volt). These values of V_T are relatively low.

The higher relative light intensity observed in S_2 device over that of S_1 device may be explained on the basis of molecular photobleaching kinetics observed in Rhodamine 6G [14]. The exact mechanism of photobleaching is not known, but it is assumed to be linked to a transition from the excited singlet state to the excited triplet state. The excited triplet state is relatively long-lived and is chemically more reactive. Moreover, Carbocyanine dye may act as an acceptor due to its high electron affinity [15]. Thus more excitons can be formed in device S_2 than device S_1 resulting in higher EL upon annihilation. On the contrary, more holes can move under the electric field in device S_1 to the cathode resulting in a large leakage current and lower V_T leading to the behavior exhibited by device S_1. The leakage current is linearly dependent on the applied voltage as indicated by the lower part of the I-V curve of S_1. Also, this leakage current is probably due to Al atoms implementation into the organic layer of S_1 during the thermal evaporation of the Al electrode. Deep implementation of Al atoms and their movement towards the other electrode would increase the leakage current and reduce the brightness of the device as the charge recombination would be less likely to occur [16].

The EL versus the applied voltage for the two samples is shown in Fig. 7. This figure reveals that the EL increases with increasing the applied voltage. At low voltages no light emission is observed. When the applied voltage exceeds V_T, light emission can be clearly observed by the naked eye in a semi-dark room. The improved performance of S_2 over that of S_1 is clearly observed. The reason for this improved performance may be attributed to the good balance in the number of injected holes and electrons in the emissive layer in S_2 compared to S_1.

The sample S_1 exhibits EL at 4.3 V, and maximum brightness is achieved using our setup at an operating voltage of 5.5 V while the sample S_2 exhibits EL at 7.9 V and maximum brightness is achieved at an operating voltage of 8.7 V.

The variation of the EL with current is depicted in Fig. 8. It is clear that the two samples exhibit almost linear relationship. A leakage current of about 3.6 mA clearly appears in the sample S_1.

Conclusions

We have conducted several measurements on two samples as OLEDs with different structures. The relative light intensity of S_2 is higher than that of S_1 but the threshold voltage for S_2 is greater than that of S_1. The maximum relative light intensity of S_1 and S_2 samples was found at 7 V and 8.7 V, respectively. Also, the value of the maximum relative intensity of the same devices was found at 10.2mA and 12.5mA, respectively. Further work should be conducted to reduce the rate of transport of electrons to the anode, as well as to reduce the rate of transport of the holes to the cathode by adding blocking layers, and/or luminescent layers, and investigating the effect of different dyes concentrations on device performance.

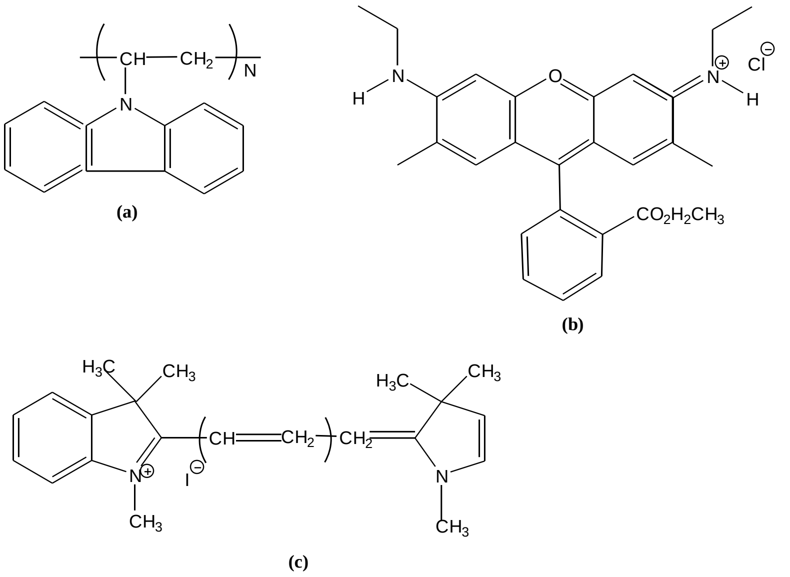

Fig. 1. The structures of (a) PVK, (b) Rhodamine 6G dye, and (c) Carbocyanine dye

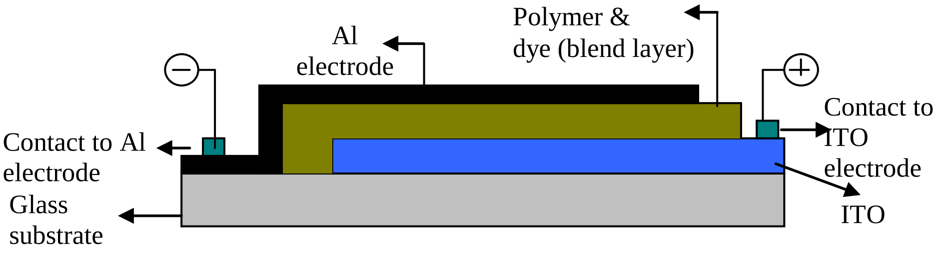

Fig. 2. A schematic diagram of the organic electroluminescent device.

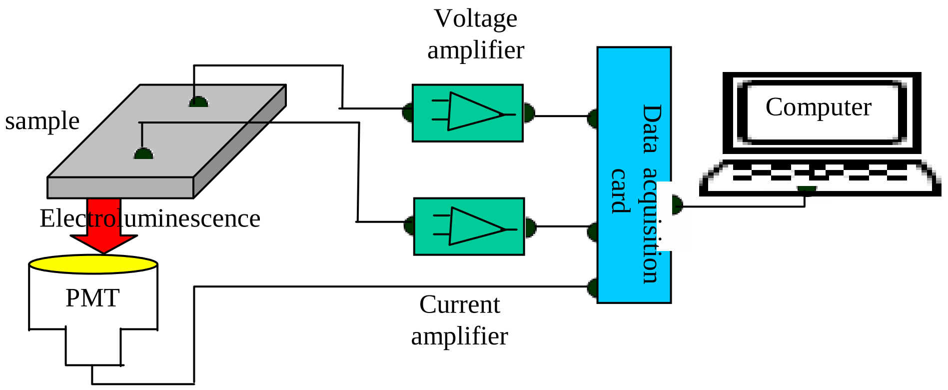

Fig. 3. A schematic diagram of the experimental setup.

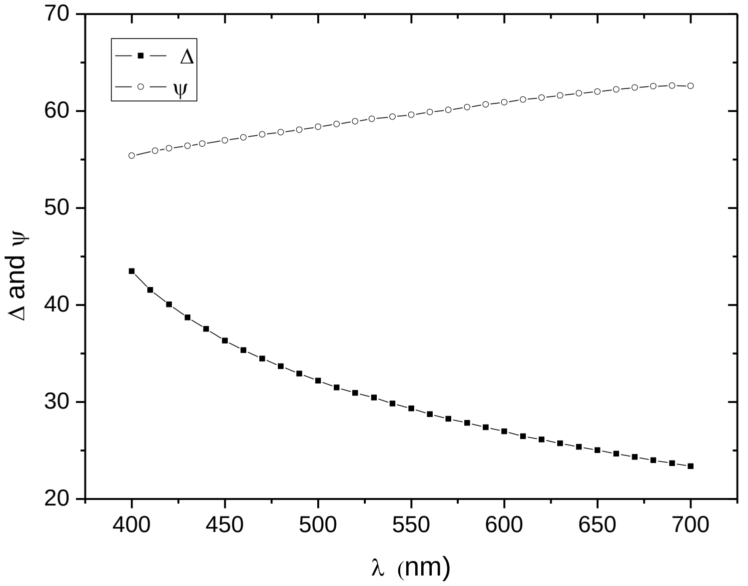

Fig. 4. ψ and Δ as a function of λ from 400 to 700 nm for 48 nm PVK film on Si wafer at 75° angle of incidence.

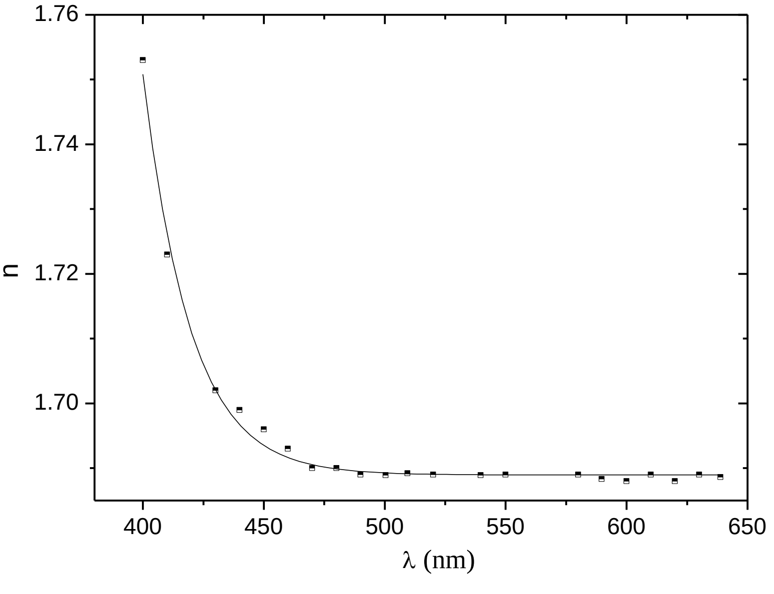

Fig.5 Variations of the refractive index n of 48 nm PVK film on Si wafer with wavelength (visible spectrum) at 75° angle of incidence.

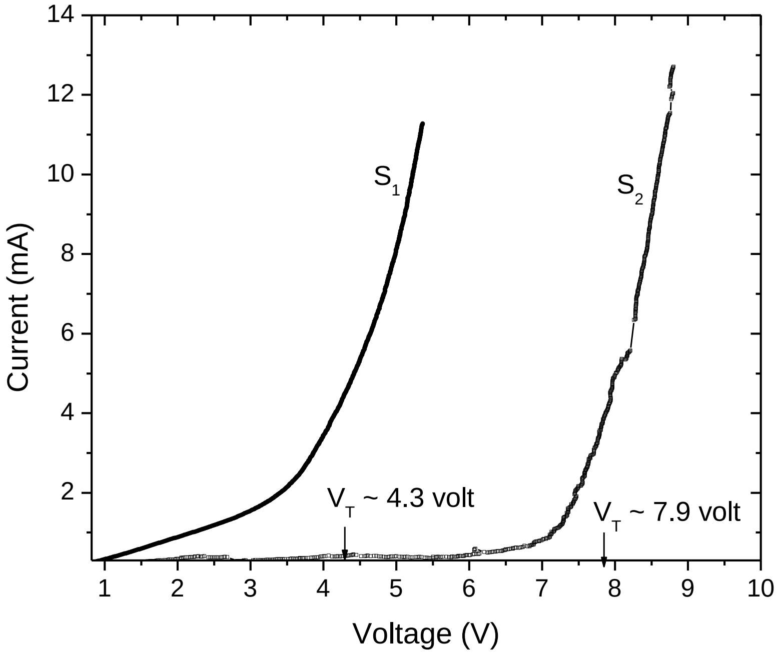

Fig. 6. I-V characteristics of ~ 48 nm devices (S1, S2). The threshold voltage V_T of each device is shown.

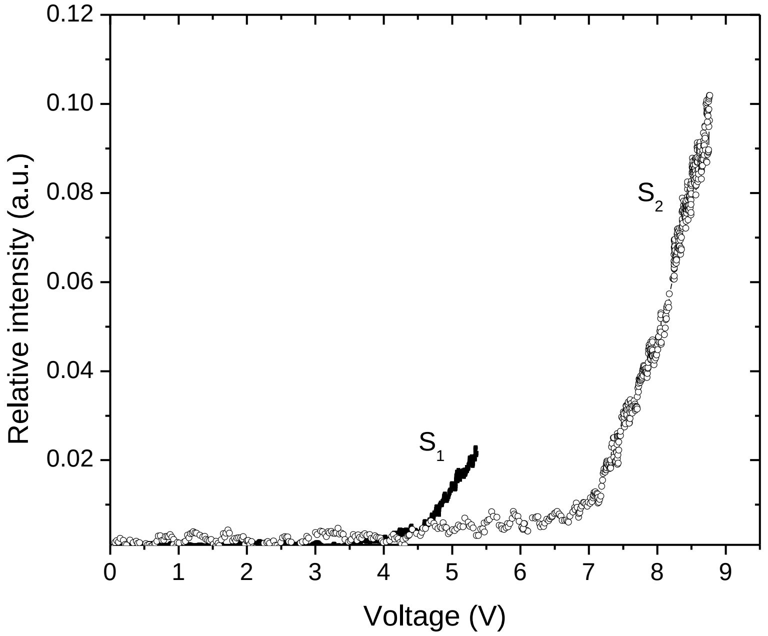

Fig. 7. Electroluminescence-voltage characteristics for S1 and S2 devices.

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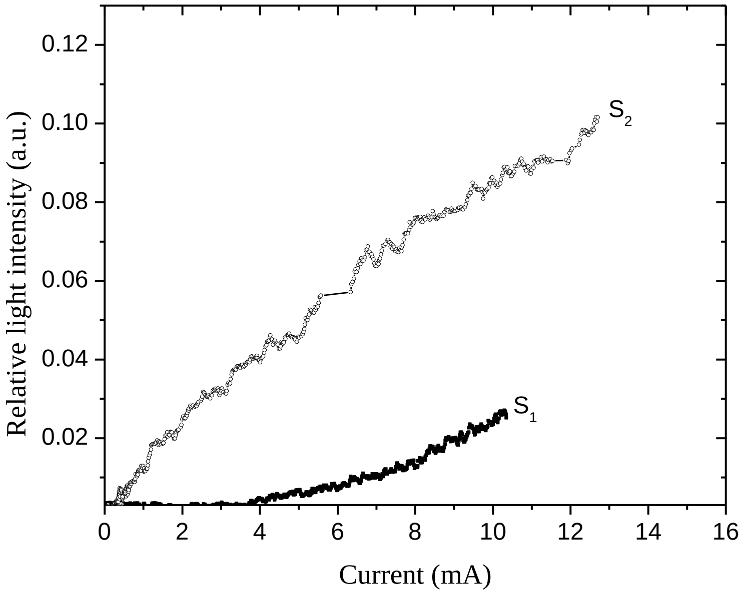

Fig. 8. The variation of EL with current for S_1 and S_2 devices.

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