ELECTROLUMINESCENCE FROM A SINGLE LAYER OF POLY(N-VINYLCARBAZOLE) DOPED WITH A NEW 1,2,4-TRIAZOLE DERIVATIVE

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Abstract: We report light emission from a single layer thin film device. This device is comprised of a hole transporting polymer, poly(N-vinylcarbazole) (PVK) doped with a new luminescent 1,2,4-triazole derivative sandwiched between an indium tin oxide electrode (ITO) and an aluminum electrode. We have studied the current versus voltage (I-V) characteristics, the electroluminescence versus voltage (EL-V) characteristics, and the lifetime of the device. Thinner samples revealed a lower EL threshold voltage $V_T$ compared with the conventional organic light emitting diodes (OLEDs) reported in the literature. This $V_T$ increased with increasing the sample thickness.

Key Words: PVK, Triazole, OLED, Electroluminescence, Ellipsometry

Introduction
Thin organic films have many attractive features and are being widely investigated for use in electronic devices. The major 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 methods used in inorganic semiconductors.

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

An OLED is a display device that sandwiches organic thin films (single or multiple layers) between two electrodes, one is a transparent indium tin oxide (ITO) anode, and one is a metallic cathode. When a voltage is applied to an OLED, electrons and holes are injected from the anode and the cathode 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(N-vinylcarbazole) (PVK) is a well known polymer for its photoconducting properties [10-12]. It, also, exhibits photoluminescent properties in the UV region [2,4,13]. In this paper, we report electroluminescence from a device composed of a single PVK layer doped with a new 1,2,4-triazole derivative namely: 3-Acetyl-1-(4-chlorophenyl)-4,5-dihydro-5,5-dimethyl-1H-1,2,4-triazole [14,15]. This material has a molecular weight of 251.72. The structures of PVK and the new triazole derivative are shown in Fig. 1.

![Fig. 1. The structures of (a) PVK and (b) the new 1,2,4-triazole derivative.](image)

**Experimental**

Each sample was arranged in an 4 x 4 array of devices. The active area of each device was 2.2 mm x 2.2 mm. Transparent conducting electrodes (ITO coated glass substrates purchased from Delta Technologies) were cut carefully into pieces of 2.5 cm x 2.5 cm. The sheet resistance $R_s$ of these electrodes was 8-12 $\Omega$/sq. Each ITO electrode was etched to form the
desired pattern by immersing in a solution of 20% HCl and 5% HNO₃ for 15 minutes at 60 °C. Then, the electrode was rinsed in DI water to remove away the leftover acids on the surface. Each ITO electrode was plasma cleaned for 8 minutes.

An amount of 300 mg of Poly(9-vinylcarbazole) PVK (Sigma-Aldrich, USA) was dissolved in 150 CC chloroform. A 2% by weight of the new 1,2,4-triazole derivative was added to the solution. The solution was then stirred thoroughly then filtered. Few drops of the solution were spun at two different speeds to form different film thickness on the etched ITO (sample 1 at 1740 rpm and sample 2 at 750 rpm). The samples were then oven heated at 80 °C for several hours under vacuum. Finally, an Al electrode was deposited by thermal vacuum evaporation. The samples were stored in dark and under vacuum.

The film thickness of the organic film was measured using a homemade variable angle spectroscopic ellipsometer VASE and found to be 48 nm and 200 nm for sample 1 and sample 2 respectively.

Electrical measurements where conducted in argon at room temperature. The driving voltages and the currents were obtained and measured using a MetraByte's Das-20 data acquisition card interfaced with a personal computer. Luminescence was detected using a photomultiplier tube (PMT). The output of the PMT was amplified then collected by the same data acquisition card. Figure 2 depicts the structure of the single layer device and the experimental setup arrangement.

![Diagram](https://via.placeholder.com/150)

**Fig. 2.** A schematic diagram of the structure of the single layer device and the experimental setup.
Results and Discussion
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. Figure 3 shows the photoluminescence and the absorption spectra of the dopant dissolved in chloroform. It illustrates that photoluminescence spectrum has a peak at 516 nm while the absorption spectrum shows a peak at 395 nm. The energies of the highest occupied molecular orbital (HOMO) and the lowest unoccupied molecular orbital (LUMO) of the triazole derivative, and the energy band gap was theoretically computed by first minimizing the energy of the compound using AM1 method and a closed shell restricted wave function. The HOMO-LUMO energies were deduced from the output of Gaussian 98 software, using hybrid density functional B3LYP and the 6-31G basis set. Results indicated that the computed HOMO and LUMO energies were 4.749 and 1.680 ev, respectively, which corresponds to a band gap energy of about 3.069 ev. The computed excitation wavelength was 404 nm as compared to 395 nm obtained experimentally.

Figure 3. The photoluminescence and absorption spectra of the new triazole derivative dissolved in chloroform.
A comparison between the current-voltage characteristics of two devices from sample 1 and sample 2 (S1 and S2) is shown in figure 4. The threshold voltages $V_T$ were estimated by linearly extrapolating the I-V curves (or EL-V curves in fig. 5) back to their intercept on the voltage axis. It was found that $V_T \sim 4.5$ V and $\sim 9$ V for S1 and S2 respectively. It is clear from this figure that a large leakage current is present in the two devices. This leakage current is linearly dependent on the applied voltage as indicated by the lower part of each IV curve. The leakage current is probably due to the Al implementation into the organic layer 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 charge recombination would be less likely to occur [16].

![Graph](image)

Figure 4. The current-voltage characteristics of S1 and S2 devices.

When the forward bias exceeds $V_T$, light emission can be clearly observed by the naked eye in a semi-dark environment. The variation of the electroluminescence EL of the device with the driving voltage is depicted in figure 5. The graph of S1 reveals EL threshold voltage $V_T$ of about 4.5 volt which is lower than the values of $V_T$ reported in the literature for conventional organic light emitting diodes (OLEDs) [3,17]. The graph of
S2 indicates an increase of \( V_T \sim 9 \) V as a result of increasing the sample thickness. A thinner polymer layer leads to a lower threshold voltage due to the high electric field injection into the polymer layer. The improved performance of S2 is clearly visible in this figure. The reason for this improved performance is probably due to the increase in the device stability by preventing shortages and sparks that might cause device failure. Moreover, increasing the thickness, within certain range, will increase the chance of excitons generation, consequently photons are emitted due to hole-electron recombination. It is clear from figure 6 that the variation of the electroluminescence with current exhibits a linear dependence.

![Figure 5](image)

Figure 5. Electroluminescence-voltage characteristics for S1 and S2 devices.
Figure 6. The variation of EL with current.

Figure 7 illustrates the variation of EL with time for S2. The EL decreases within the first 200 minutes then it saturates with some EL variations. The device exhibits a lifetime of about 680 minutes. Visual inspection of the device after measurements revealed that the reason for the device failure is the degradation of the Al contact at the sharp edges of the device which resulted in an open circuit.
The lower performance observed in our single layer devices compared with those reported in literature may be explained as follows: A thin single emitting layer device exacerbates two common problems. The first is that the roughness of the ITO may introduce peaks that vary in height. This creates a non-uniform interface between the ITO electrode and the organic layer. Areas where the peaks penetrate the organic layer are thinner than areas without peaks which provide thinner pathways with lower resistance path for charge transport. Therefore, the current densities in these regions are increased. When the applied voltage is increased, these small pathways are overloaded and burn out. If these regions are large enough then the whole device can be damaged and destroyed. The second problem is that the charge balance in the emitting layer is necessary to produce a bright and efficient device. If only one type of charge carriers exists in the sample then the charge recombination can not occur. The thin layer in these devices provides only limited time for charge carriers recombination before reaching the other electrode (see fig.5). Moreover, the recombination zone is more likely to be close to the metal electrode increasing the probability that the light emission will be quenched by the metallic Al cathode [18]. Electrode polarization may contribute to the decay of the EL with time, as the EL output was observed to improve after the bias was stopped for sometime.
Conclusion
We report electroluminescence from devices composed of a single PVK layer doped with a new 1,2,4-triazole derivative sandwiched between transparent ITO and Al electrodes. Thinner samples showed a lower EL threshold voltage $V_T$ compared with the conventional organic light emitting diodes (OLEDs) reported in the literature. This $V_T$ increased with increasing the sample thickness. The device EL performance increases with increasing the thickness. More work is needed to improve the performance by incorporating electron transport media and buffer layers.

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References