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Abstract: In this work, organic light emitting diodes were fabricated using a double layer device with the general structure ITO/PVK/dye/InGa. Three different dyes were used, namely: Rhodamine B, Crystal violet, and Bromophenol blue. Three different PVK layer thicknesses were used with each dye. The double layer system was sandwiched between indium tin oxide (ITO) and Indium-Gallium alloy (InGa) as anode and cathode, respectively. The J-V characteristic curves were investigated in details with the PVK layer thickness. The conduction mechanisms were also studied.

Keywords: PVK, organic light emitting diode, electroluminescence, SCLC, Mott Gurney Law, Quasi ohmic

1. INTRODUCTION

Organic light emitting diode (OLED) is a thin-film optoelectronic device consisting of a single layer, double layer or multilayer of organic materials sandwiched between two electrodes, at least one of which is transparent or semi-transparent for the transmission of light. Organic light emitting diodes have been the focus of intense study since the late 1980s, when the low voltage organic electroluminescence in small organic molecules such as Alq₃, and large organic molecules such as polymers (PPV), were reported [1, 2]. Since that time, research has continued to demonstrate the potential of OLEDs as viable systems for displays and eco-friendly lighting applications. The recent rapid development of organic light-emitting diodes (OLEDs) has resulted in the commercialization of simple dot-matrix OLED displays. The great success of OLED devices has also introduced many new organic semiconductors. From a fundamental perspective, these devices work by injection of charge carriers (holes and electrons) from metal electrodes into organic semiconducting layers which transport through the device and recombine to form excited states (excitons) that emit light upon relaxation. Many OLED displays have been commercialized and now, the researchers are trying hard to commercialize the OLED-based solid state lighting devices.

Organic materials have previously been considered for the fabrication of electroluminescent devices. The primary reason is that a large number of organic materials are known to have extremely high florescence quantum efficiencies in visible spectrum, including blue region. The first observations of electroluminescence (EL) in organic materials were made in the early 1950s by Andre Bernanose and co-workers [3-5] at the Nancy University, France. They applied high voltage alternating current fields in air to materials such as acridine orange, either deposited on or dissolved in cellulose or cellophane thin films. The proposed mechanism was either direct excitation of the dye molecules or excitation of electrons. Although they insisted on the similar excitation and emission mechanisms that had been established in inorganic EL in those days, it was understood by Short and Hercules et al. [6] that the emission was induced from the secondary ultraviolet light by a glow discharge between two electrodes.

In the 1960s, research moved towards the carrier-injection type of electroluminescence, namely OLED, in which a highly purified condensed aromatic single crystal, especially an anthracene was used. Martin Pope and co-workers at New York University [7, 8] and W. Helfrich and Schneider [9, 10], in particular, performed experiment related to carrier recombination and the emission mechanism, and the physical interpretation proposed by them is still considered very useful today. While a highly purified zone-

refined anthracene single crystal essentially shows a conductivity of 10^{-20} S/cm, double injection of holes and electrons were achieved efficiently which was based on space-charge-limited current (SCLC) with the equipment of charge-carrier-injection electrodes, and such experiment resulted in a successive carrier recombination, the creation of singlet and triplet excitons, and the radiative decay of them. In this way, the basic EL process has been established since the 1960s. Pope's group also first observed DC electroluminescence under vacuum on a pure single crystal of anthracene and on anthracene crystals doped with tetracene in 1963 using a small area silver electrode at 400V. The proposed mechanism was field-accelerated electron excitation of molecular fluorescence.

In 1987 the first organic light emitting diode (OLED) was fabricated by Tang and VanSlyke [1]. An OLED is a semi conducting device which consists of small molecule organic material thin films that are sandwiched between two conducting thin films. OLEDs emit light when a proper voltage is applied to it. There has been an interest [11-18] in the study of OLEDs because of their low operating voltages, structure flexibility and low fabrication cost. The efficiency of an OLED depends on its structure which includes polymer materials and dyes and on the design of the device. In general, multi-layer devices exhibit better performance than single-layer devices because the injection and recombination of holes and electrons are better balanced. The recombination of electrons and holes is called exciton which occurs into the emitter layer.

Recently, there has been a wide interest in the use of organic polymers in OLEDs manufacturing due to their potential characteristics, the simple physical manipulation, and the great flexibility of chemical structures. Polymers allow the development of innovative and low cost devices where flexible, large area, and transparent substrates can be used. Moreover, the simple processing and deposition techniques of polymers make them strong candidates for many applications. Poly N-vinyl carbazole (PVK) is the most interesting material in this field because of its remarkable photoconductivity properties. The electrical properties with its chemical and thermal resistance make the PVK useful in the electronics industry. PVK is considered a hole transport material, i.e., the hole mobility is much greater than electron mobility. PVK has been widely utilized in the fabrication of OLEDs.

A frequently used method to analyze charge carrier transport in organic semiconductors is based on space-charge-limited current measurements performed on single carrier devices [11-24]. It is to be noted that SCLC dominates at high current, where traps are filled and the electrode/organic contact is Ohmic or quasi-Ohmic. In the work reported by Mori et al. [25], SCLC, however, was not observed in multilayer small molecular OLEDs. It was not only attributed to failure of the Ohmic contact presumption at the electrode/organic interface, but also to the energy barriers between the organic layers. On the other hand, the SCLC model is easily accepted in much simpler structures, such as polymer LEDs [26] and single layer hole-only small molecular devices [27]. If the electron-only device were, however, undoped and the injection at the contacts efficient, the current density *J* would to a first approximation not depend on the equilibrium electron concentration anymore. Instead *J* would just depend on the mobility μ , which is typically the only unknown parameter, as well as the voltage *V*, the device thickness *d* and the permittivity $\varepsilon = \varepsilon_0 \varepsilon_r$ and would ideally follow the Mott–Gurney law [28,29]

$$J = \frac{9}{8}\varepsilon_0\varepsilon_r \mu \frac{V^2}{d^3}$$
(1)

Here ε_0 is the vacuum permittivity and ε_r is the relative permittivity. The Mott–Gurney law is frequently used to determine the mobility of organic semiconductors used for light emitting diodes and solar cells. However, its derivation uses three assumptions that are often not applicable in organic semiconductors, namely that the device is trap-free, that diffusion is negligible and that the electric field at the injecting contact is zero. All three assumptions are in general not correct, in particular the assumption that there are no charged defects in organic semiconductors [30-36]. While there have been numerous attempts to develop models to take traps in unipolar devices into account [37-41], nearly all of them still rely on drift as the only transport mechanism. However, traps will often lead to a situation where diffusion currents cannot be neglected anymore, which makes analytical approximations for this situation problematic [17,20, 23].

In this paper, we report electroluminescence from a double layer device composed of a dye layer deposited on PVK layer. Three different dyes were used namely: Rhodamine B ($C_{28}H_{31}C_1N_2O_3$), Crystal

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violet ($C_{25}N_3H_{30}Cl$), and Bromophenol blue ($C_{19}H_{10}Br_4O_5S$). The effect of changing the PVK layer thickness on the J-V characteristic curves and on the electroluminescence emitted from the samples was investigated. The thickness of the PVK layer was measured using a home-made ellipsometer.

The structures of PVK and the three organic dyes are shown in Fig. 1.



Fig1. The structures of (a) PVK, (b) Rhodamine B, (c) Crystal violet and (d) Bromophenol blue.

Figure2 shows a schematic diagram of the double layer device (ITO/PVK/dye /InGa)



Fig2. Schematic drawing of a double layer device

Table1.	Different	weights	of the	PVK	polvmer	used to	o construct	the PVK	laver.	(Ref.	Taher.M	1 et al.)
										()		_

Sample No.	Polymer	Weight	Solvents	Thickness(nm)
S1	PVK	10mg	5mlTHF+1ml	35.33
		-	Toluene	
S2	PVK	15mg	5mlTHF+1ml	44.30
		-	Toluene	
S3	PVK	20mg	5mlTHF+1ml	94.48
		C	Toluene	

Fable2. The structure of dou	ble layer device.	.(Ref. [Taher.M] et al.)
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Group No.	Structure	Sample No.	Thickness of	Thickness of
			PVK (nm)	dye (nm)
G1	ITO/PVK/Rhodamine B/InGa	G1S1	35.33	50
		G1S2	44.30	
		G1S3	94.48	
G2	ITO/PVK/Crystal Violet/InGa	G2S1	35.33	50
		G2S2	44.30	
		G2S3	94.48	
G3	ITO/PVK/Bromophenol	G3S1	35.33	50
	blue/InGa	G3S2	44.30	

	G3S3	94.48	

The conduction mechanism of regions with slopes very close to unity is said to be ohmic. In regions with slopes equal to 2, the conduction mechanism is called space charge limited [40, 41] in which J and V obey the relation

$$J_{SCL} = \frac{9}{8} \mu \varepsilon \frac{V^2}{d^3}$$



Fig3. Characteristics curve in logarithmic scale for ITO/35.33nm PVK/50nm Rhodamine B/InGa device.



Fig4. Characteristics curve in logarithmic scale for ITO/44.3nm PVK/50nm Rhodamine B/InGa device.

In figures 3 and 4, at low voltages the lnJ -lnV characteristic curves show slopes for region AB to be very close to unity in many samples. This means that the conduction in this region follow Ohm's law which is given by the equation

$$J_{ohm} = qn\mu \frac{V}{d}$$
(3)

where q is the electronic charge, n is the charge carriers density, μ is the carriers mobility and d is the thickness

2. DERIVATION OF EXPRESSIONS CORRELATING CURRENT AND VOLTAGE OF OLEDS

2.1. Space Charge Limited Current

In modern OLEDs [43], standard electroluminescence operation requires current injection >3 mA/cm². However, the carrier mobility is low, being 10^{-6} - 10^{-4} cm²/(V.s) for electrons and 10^{-5} - 10^{-3} cm²/(V.s) for holes. Such strong injection into low mobility materials inevitably leads to charge accumulation in organic materials. In this case, the I-V relation follows Mott-Gurney relation, also known as Child's law, for trap-free unipolar conduction. If the contact between the metal and the organic semiconductor is Ohmic with a contact resistance much lower than the resistance of the bulk material, then in this case, the current is easily injected into the organic material and the transport of charge is dominated by the bulk [44]. In fact, by Ohmic contact we mean that the electrode is an infinite reservoir of charge, which

(2)

can maintain a steady state space-charge limited current (SCLC) in the device [45]. In contrast, in the case where the injected charge dramatically changes the electric field configuration in the materials, that is, effectively screens the source-drain field, and the transport becomes space-charge limited. In this case, the I-V curves look linear if the field due to the applied bias is the dominant electric-field in the device. In this case, the conduction is usually linear in the low source-drain bias because the current density in the materials is low. On the other hand, at higher fields where the current density is very high, there is a higher concentration of charge carriers in transit between the source and the drain. Consequently, the screening due to these "space charges" gives rise to nonlinear I-V characteristics. It is to be noted that space-charge effects are more readily maintained in organic semiconductors where the mobility is poor. In fact, the low mobility greatly prevents the collection of the carriers at the drain or the recombination of opposite charges. Furthermore, organic semiconductors possess a large concentration of highly localized states (i.e., traps, defects, etc.) which can trap mobile charge carriers temporarily or permanently (immobile charges) [46]. Thus, all these factors make organic semiconductors the perfect breeding ground for space-charge transport. For obtaining a current-voltage relation for samples operating in the space-charge limited currents regime, let us use the current density given by the equation

$$j = \mu q \rho(x) F(x) \tag{4}$$

where μ is the mobility defined as the constant of proportionality in the relationship between the applied electric field F(x), and the drift velocity of the carriers, v_d , given by $v_d = \mu F$, q is the charge of the electron, $\rho(x)$ is the total density of carriers given by, $\rho(x) = \rho_c(x) + \rho_t(x)$, where $\rho_c(x)$ is the density of carriers in conductive states, and $\rho_t(x)$ is the density of carriers in trapped states.

Using Poisson's equation, we can write the following relation

$$\frac{dF}{dx}\frac{k\varepsilon_0}{q} = \rho(x) \tag{5}$$

From Equs. (1) and (2), we get

$$j = \mu q \frac{dF}{dx} \frac{k\varepsilon_0}{q} F(x)$$

Or,

$$j = \mu \frac{dF}{dx} k \varepsilon_0 F(x) \tag{6}$$

where, k is the dielectric constant of the semiconductor materials, and \mathcal{E}_0 is the permittivity of the free space. The integration of both sides of Eq. (3) gives

$$\int_{0}^{x} j dx = \int_{0}^{x} \mu \frac{dF}{dx} k \varepsilon_{0} F(x) dx$$
Or,
$$\int_{0}^{x} j dx = \int_{0}^{x} \mu k \varepsilon_{0} F(x) dF$$
Or,
$$jx = \mu k \varepsilon_{0} \int_{0}^{x} F(x) dx$$
Or,
Or,

 $jx = \mu k \varepsilon_0 \left[\frac{F(x)^2}{2} \right]$

Or,

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$$jx = \mu k \varepsilon_0 \left[\frac{F(x)^2}{2} - \frac{F(0)^2}{2} \right]$$
(7)

In the case of Ohmic contacts, the electric field at the interface (x = 0) should be zero, that is,

$$F(0) = 0$$
. Thus, we get $jx = \mu k \varepsilon_0 \left[\frac{F(x)^2}{2} \right]$

Or,

$$2jx = \mu k\varepsilon_0 F(x)^2$$

Or,

$$\frac{2jx}{\mu k\varepsilon_0} = F(x)^2$$

Or,

$$F(x) = \sqrt{\frac{2jx}{\mu k\varepsilon_0}}$$

(8)

Now using
$$V = \int_{0}^{L} F(x) dx$$

Eq. (2) can be expressed as

$$V = \int_{0}^{L} \left[\frac{2jx}{\mu k \varepsilon_0} \right]^{\frac{1}{2}} dx$$

Or,

$$V = \left[\frac{2j}{\mu k\varepsilon_0}\right]^{\frac{1}{2}} \int_0^L (x)^{\frac{1}{2}} dx$$

Or,

$$V = \left[\frac{2j}{\mu k \varepsilon_0}\right]^{\frac{1}{2}} \left[\frac{x^{\frac{3}{2}}}{\frac{3}{2}}\right]_0^L$$
$$V = \left[\frac{2j}{\mu k \varepsilon_0}\right]^{\frac{1}{2}} \frac{L^{\frac{3}{2}}}{\frac{3}{2}}$$
$$V = \left[\frac{2j}{\mu k \varepsilon_0}\right]^{\frac{1}{2}} \frac{2}{3} L^{\frac{3}{2}}$$

Squaring both sides of Eq. (9), we get

$$V^2 = \frac{2j}{\mu k\varepsilon_0} \frac{4}{9} L^3$$

Or,

$$\frac{V^2}{L^3} = \frac{8j}{9\mu k\varepsilon_0}$$

Or,

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(9)

$$j = \frac{9}{8} \mu k \varepsilon_0 \frac{V^2}{L^3}$$

(10)

3. CHARACTERISTIC CURVES

Figures 5, 6 and 7 illustrate the J-V characteristic curve for groups G1, G2 and G3 respectively. The J-V characteristic curves show an exponential increase of current with the applied voltage. Under forward bias the current density increases slowly with increasing voltage. At certain higher voltage (threshold voltage V_{th}) the current density increases sharply. The threshold voltage is the voltage at which an OLED begins to emit light. The threshold voltage can be estimated from the relative light intensity-voltage characteristic curve. To study the effect of increasing thickness of the PVK layer the J-V curves of all samples of group G1 are shown together in Fig. 5 whereas for all sample of group G2 in Fig. 6 and for all samples of group G3 in Fig. 7, they show that , as the thickness increases the sample exhibit more resistance to the current flowing through it and the current density decreases for the same voltage.



Fig5. J-V curves of all samples of ITO/PVK/50nm Rhodamine B/InGa.



Fig6. J-V curves of all samples of ITO/PVK/50nm Crystal violet/InGa.



Fig7. J-V curves of all samples of ITO/PVK/50nm Bromophenol blue/InGa

Figure 8 shows the energy band diagram of the ITO/PVK/Rhodamine B/ InGa sample. The figure shows the work functions of the electrodes as well as highest occupied molecular orbitals (HOMO), and lowest unoccupied molecular orbital (LUMO) of PVK and Rhodamine B [42].



Fig8. Mechanism of electroluminescence of a ITO/PVK /Rhodamine B/InGa device

4. CONCLUSION

We have investigated the electroluminescence from devices composed of a PVK layer doped with Rhodamine B, crystal violet, and bromophenol blue dyes. The fabricated devices are composed of a double layer with the general structure ITO/ PVK/dye/ InGa. The electroluminescence emitted from these devices has been investigated with applied voltage. The J-V characteristics showed an exponential increase of current with the applied voltage in most samples, which is similar to a typical diode characteristic curve. It also shows clearly that no electro luminescence can be detected at low applied voltage. The samples ITO/35.33nm PVK/50nm Rhodamine B /InGa and ITO/44.3nm PVK/50nm Rhodamine B /InGa were found to be organic light emitting diodes. The results include the current-voltage characteristics and current-voltage relation in logarithmic scale.

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