Mobility and Traps Effect in Semiconducting Polymer by Mobility Models

Muhammad Ammar Khan[*], Ahmad Neyaz Khan[†], Amr S. Zalhaf[‡]

Abstract

We apply three different mobility models named exponential model of Pai, mobility edge model with single Gaussian DOS, and double Gauss model to one organic diode with polymer poly(3-hexylthiophene) (P3HT). These three models are different from one another with respect to their function. The results obtained from three different models are well explained in this paper by acquiring the suitable parameters, and gives satisfactory results. When the parameter and results of these models are compared with one another the modified double Gauss model agrees well with the current -voltage data. While exponential model of Pai and mobility edge model with single Gaussian DOS shows the abnormality condition in mobility range.

***Keywords*:** organic diode, non-symmetric barriers, Gauss traps, Einstein relationship

Introduction

Organic semiconductors are the next generation materials for the fabrication of future electronic devices with promise of radio-frequency identification (RFID) tags and active matrix display (Arias et al., 2010)(Chabinyc et al., 2005)(Katz & Huang, 2009)(Kelley et al., 2004)(Sirringhaus, 2009)(Khokhar et al., 2020). The mobility have been achieved in thin film transistors with a value of 1.4 cm^2/Vs for polymers (Tsao et al., 2009), 3.4 cm^2/Vs for polycrystalline films(Kelley et al., 2003), and 30 cm^2/Vs for

[*]Department of Physical and Numerical Sciences, Qurtuba University of Science & Information Technology, D. I. Khan, Pakistan, ammar@qurtuba.edu.pk,, PhD, University of Electronic Science and Technology of China, Chengdu, P. R. China.

[†]Department of Computer Application, Integral University, Lucknow, India, ahmadnk500@gmail.com, University of Electronic Science and Technology of China, Chengdu, P. R. China.

[‡]Electrical Power and Machines Engineering Department, Tanta University, Tanta 31511, Egypt, amr.salah@f-eng.tanta.edu.eg, University of Electronic Science and Technology of China, Chengdu, P. R. China.

single-crystal-like films of small molecule organic semiconductors (Minemawari et al., 2011).

(Mesta et al., 2013), and (Khan & Jiu-Xun, 2015)(Khan, Sun, et al., 2015) pointed out that since many mobility models have been proposed, the researches on transport mechanism(Romele et al., 2021)(Li et al., 2021) under direct current conditions have been matured, and more attention should be paid to properties at alternating conditions. We notice that one mobility model has been applied to some materials and other mobility model is applied to some other materials, it is not sure that whether the one mobility model is applicable to all other organic semiconductors or not, and one organic polymer is suitable for every mobility model or not. So, it is necessary to apply every mobility model to more and more materials to check their applicability, and judge whether we need to combine some models together or propose new models.

The trap states' effect is observed in organic semiconductors(Xie et al., 2009)(Völkel et al., 2002)(Verlaak et al., 2003). with characterization of accurate distributions and their relationship with defects is fully characterized the materials limits. Moreover, it is observed that single-crystal studies represent the performance of organic semiconductors with higher mobility which is observed in (Calhoun et al., 2007)(Nicolai et al., 2011).

The traps effect is caused by the residual impurities, and lattice disorder of organic semiconductors. Transport models are the theoretical approach to estimate the trap distribution with characteristics of direct fitting of space charge limited current (SCLC)(Xie et al., 2009)(Krellner et al., 2007), but nowadays the most practicable distribution for density of state (DOS) is the Gaussian distribution.

Ammar et.al modified, and developed some mobility models(Khan, Jiu-Xun, et al., 2015)(Khan & Jiu-Xun, 2015)(Khan & Sun, 2015) and pointed out the neutral condition(Khan, Sun, et al., 2015) applied to different organic diodes, an excellent agreement is obtained between theoretical results and experimental data. In our recent work(Ammar Khan et al., 2020) the transport model of improved J-V formula based(Khan & Jiu-Xun, 2015) is applied to hole only devices, the results verified that the modified charge transport model is accurate, and precise.

Now in this work, we consider three different models; the exponential model of Pai (Pai et al., 1984), double Gauss model(Nicolai et al., 2011), and mobility edge model with single Gauss DOS(Dacuña & Salleo, 2011) to apply on poly(3-hexylthiophene)(P3HT), the obtained results are very good.

Method and Models Description

The Space charge limited characteristic is described by the perturbation of following equation (1).

$$\frac{d^2\varphi}{dx^2} = -\frac{q}{\varepsilon_r \varepsilon_0} p(x) \tag{1}$$

In above Eq. (1) φ is the electric field, x is the coordination, q is the elementary charge, p is density of hole; ε_0 is the free-space permittivity, ε_r is the dielectric constant of the semiconductor.

The above equation with trapped charges can be written as

$$\frac{d^2\varphi}{dx^2} = -\frac{q}{\varepsilon_r \varepsilon_0}(p_f + p_t) \tag{2}$$

Here we apply the drift-diffusion equation for diffusion current with holes mobility μ_p

$$J(x) = -q\mu_p(x)p(x)\frac{\partial\varphi}{\partial x} - kT\mu_p(x)\frac{\partial p(x)}{\partial x} \tag{3}$$

We Assume that the thickness of organic layer is L. Take ($x = 0$) W_{left} for contact on the left-side low potential, and ($x = L$) W_{right} for contact on the right-side high potential.

The boundary conditions with drop electric potential V can be expressed as below

$$\varphi(0) = W_{left} + V\ , \quad \varphi(L) = W_{right} \tag{4}$$

$$V - V_{bi} = \varphi(0) - \varphi(L) \tag{5}$$

The general equation of the Gaussian model for DOS with the total number of states N_0, the center energy E_v is given below

$$D(E) = \left(N_0 / \sigma\sqrt{2\pi}\right)\exp\left[-(E - E_v)^2 / 2\sigma^2\right] \tag{6}$$

Where σ is the standard deviation of the Gaussian distribution. In case of degenerate organic semiconductors, the density of hole can be expressed as

$$p = \int_{-\infty}^{\infty} \frac{D(E)}{1 + \exp[(E_F - E + q\varphi(x))/kT]} dE \tag{7}$$

In the above equation the Fermi-Dirac statistics represent the following generalized Einstein relation (GER) with diffusion coefficient of holes D_p.

$$D_p / \mu = -p\left[q dp / dE_F\right]^{-1} \tag{8}$$

In organic semiconductors the Einstein relation for single-carrier diodes is valid for diffusion-driven currents.
The Fermi-Dirac statistics in Eq. (6) can be replaced by the Boltzmann statistics, Eq. (7) is changed as

$$p = \int_{-\infty}^{\infty} D(E)\exp[(E - E_F - q\varphi(x))/kT]dE \tag{9}$$

Substituting Eq. (5) into Eq. (8), introducing dimensionless variable, $y = (E - E_v)/\sigma\sqrt{2}$, we can evaluate the integral

$$\int_{-\infty}^{\infty} \exp\left[-(E - E_v)^2/2\sigma^2\right]\exp[(E - E_v)/kT]dE = \sigma\sqrt{2\pi}\exp\left[\sigma^2 / \left(\sqrt{2}kT\right)^2\right]$$

And the hole density can be expressed as

$$p = N_f \exp\left[-q\varphi(x)/kT\right] \tag{10}$$

with effective DOS represents

$$N_f = N_0 \exp[(E_v - E_F)/kT]\exp\left[\sigma^2/2(kT)^2\right] \tag{11}$$

The mobility models are used for solving the drift-diffusion equation (3)

In previous work, we have considered two mobility models but here we consider three models; exponential model of Pai, mobility edge model with single Gaussian DOS, and double Gauss model to apply on P3HT. Mobility is expressed as function $\mu(T, p, E)$.

$$\mu_p(E) = \mu_p(0)\exp(\gamma\sqrt{E}) \tag{12}$$

where $\mu_p(0)$ zero field mobility. The charge carrier's mobility μ with an electric field E express the dependence of $\mu_p(E)$ on $\sqrt{E}$ with random distribution.

The observation between γ and $1/T$ can be expressed as following relations.(Blom et al., 1997)

$$\mu_p(0) = \mu_0 \exp(-\Delta/k_B T) \tag{13}$$

$$\gamma = B\left(\frac{1}{k_B T} - \frac{1}{k_B T_0}\right) \tag{14}$$

we find that Eqs. (13) and (14) are not accurate enough, and we extend Eqs. (13) and (14) to following forms

$$\mu_p(0) = \mu_0 \exp\left[\Delta_1/(kT) + \Delta_2/(kT)^2\right] \tag{15}$$

$$\gamma = \gamma_0\left[1 + \Delta_3/(kT) + \Delta_4/(kT)^2\right] \tag{16}$$

with six parameters μ_0, Δ_1, Δ_2, γ_0, Δ_3, Δ_4.

The second model which has considered in this paper is the mobility edge model with single Gaussian DOS(Dacuña & Salleo, 2011). In this model we express the free carriers and also a trapped carriers with widths σ, center energy levels E_v, respectively as shown in Eq

(6). Following equations represents the densities of free holes, and trapped holes

$$p_f = \int_{-\infty}^{E_m} D(E) f(E) dE \tag{17}$$

$$p_t = \int_{E_m}^{\infty} D(E) f(E) dE \tag{18}$$

Here E_m is the mobility edge, electronic states with higher energy.

$$f(E) = \{1 + \exp[(E_F - E + q\varphi(x))/k_B T]\}^{-1} \equiv \{1 + \exp[(E_F(x) - E)/k_B T]\}^{-1} \tag{19}$$

Where $f(E)$ is the Fermi-Dirac (FD) distribution, E_F Fermi energy level, and $E_F(x) = E_F + q\varphi(x)$ is the quisi-Fermi energy level.

Under the space charge limited conditions, the equation for free hole (Eq. (17)) can be simplified in term of Eq (9) as following form

$$p_f = \int_{-\infty}^{E_m} D(E) \exp[(E - E_F - q\varphi(x))/kT] dE \tag{20}$$

With some necessary modification of eq (11), effective DOS can be defined as

$$N_f = N_0 \exp[(E_v - E_F)/kT] \cdot \int_{-\infty}^{E_m} \exp[-(E - E_v)^2/2\sigma^2] \exp[(E - E_v)/kT] dE \tag{21}$$

By solving the Fermi energy E_F from Eqs. (10) and substituting it into Eq. (19), we obtain

$$f(E) = \{1 + \exp[(E_F - E + q\varphi(x))/k_B T]\}^{-1} = \{1 + (N_f / p_f) \exp[(E_F - E)/k_B T]\}^{-1} \tag{22}$$

By substituting above Eq. (22) and Eq. (6) into Eq. (18) then we get following relation

$$p_t = N_0 \int_{E_m}^{\infty} \exp[-(E - E_v)^2/2\sigma^2] \{1 + (N_f / p_f) \exp[(E_F - E)/k_B T]\}^{-1} dE \tag{23}$$

Moreover, the Fermi distribution is applicable to free and trapped holes. However, the Boltzmann distribution is used for free holes.

We derive following relation from trap hole Eq. (22)

$$p_f (\partial f / \partial p_f) = \{1 + (N_v / p_f) \exp[(E_F - E)/k_B T]\}^{-2} (N_f / p_f) \exp[(E_F - E)/k_B T] \tag{24}$$

Substitution of Eq. (22) into Eq. (24) we get

$$p_f\left(\partial f/\partial p_f\right)= f^2\left(f^{-1}-1\right)= f(1-f) \tag{25}$$

The derivatives of trapped charges with respect to the free charges is expressed in the following form

$$p_f\left(\partial p_t/\partial p_f\right)=\left(N_0/\sigma\sqrt{2\pi}\right)\int_{E_m}^{\infty}\exp\left\{-\left[(E-E_v)/\sigma\sqrt{2}\right]^2\right\}f(E)[1-f(E)]dE \tag{26}$$

The third model we have the considered energy-band model for p-type material. The model consists two Gaussian DOS (free carriers DOS, trapped carriers DOS). We can write Eq. (6) as following form

$$D_f(E)=\left(N_0/\sigma\sqrt{2\pi}\right)\exp\left[-(E-E_v)^2/2\sigma^2\right] \tag{27}$$

$$D_t(E)=\left(N_t/\sigma_t\sqrt{2\pi}\right)\exp\left[-(E-E_t)^2/2\sigma_t^2\right] \tag{28}$$

The densities of holes (free and trapped) in term of double Gauss model represent by the Eq.(17) and Eq.(18) by taking the complete Gaussian integral range from –∞ to ∞.

$$p_f=\int_{-\infty}^{\infty}D_f(E)f(E)dE \tag{29}$$

$$p_t=\int_{-\infty}^{\infty}D_t(E)f(E)dE \tag{30}$$

And Eq. (29) for free holes can be simplified in term of Eq. (9) as following form

$$p_f=\int_{-\infty}^{\infty}D_f(E)\exp[(E-E_F)/kT]dE \tag{31}$$

Substituting Eq. (27) into Eq. (19) we get the integral

$$\int_{-\infty}^{\infty}\exp\left[-\left(E-E_v\right)^2/2\sigma^2\right]\exp[(E-E_v)/kT]dE=\sigma\sqrt{2\pi}\exp\left[\sigma^2/\left(\sqrt{2}kT\right)^2\right] \tag{32}$$

Now the equation of derivation of trapped charges with respect to the free charges is

$$p_f\left(\partial p_t/\partial p_f\right)=\left(N_t/\sigma_t\sqrt{2\pi}\right)\int_{-\infty}^{\infty}\exp\left\{-\left[(E-E_t)/\sigma_t\sqrt{2}\right]^2\right\}f(E)[1-f(E)]dE \tag{33}$$

Let trapped charges do not contribute to current, so drift-diffusion equation for current in for Eq. (3) can be written into following way

$$J=-q\mu p_f\frac{\partial\varphi}{\partial x}-kT\mu\frac{\partial p_f}{\partial x} \tag{34}$$

Eq. (34) is the drift-diffusion equation for current refers to mobility edge model with single DOS, and double Gauss model. while Eq. (3) is the drift-diffusion equation for current refers to exponential model. Considering the E_t (single trap level) the density of trapped charges in term of Eqs. (18), (22) can be written as

$$p_t=N_0\int_{E_m}^{\infty}\exp\left[-\left(E-E_v\right)^2/2\sigma^2\right]\left\{1+\left(N_f/p_f\right)\exp\left[\left(E_F-E\right)/k_BT\right]\right\}^{-1}dE \tag{35}$$

Comparison of Results

Now we apply above three models; which are exponential model of Pai, single Gaussian DOS mobility edge model, and double Gauss model to P3HT poly(3-hexylthiophene), with 95nm thickness 95nm. In figures 1-3, we have calculated the J-V curves for the P3HT material and compared results with experimental points. These figures show that the comparison of theoretical results with experimental points is fine.

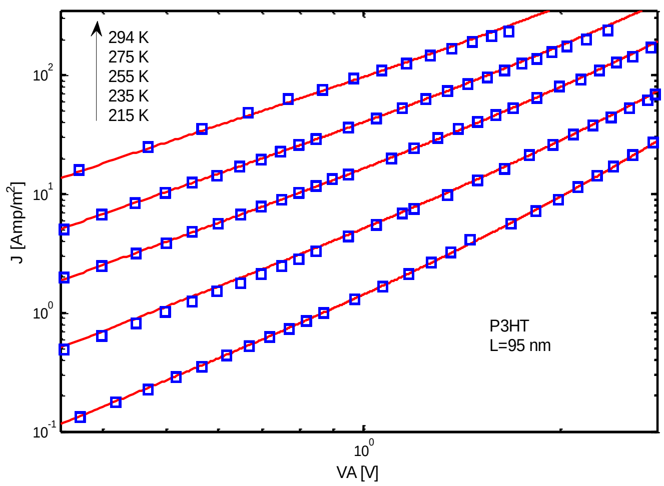

Fig.1 (color online). By applying exponential model; (symbols represent experimental data, solid lines represent theoretical results)

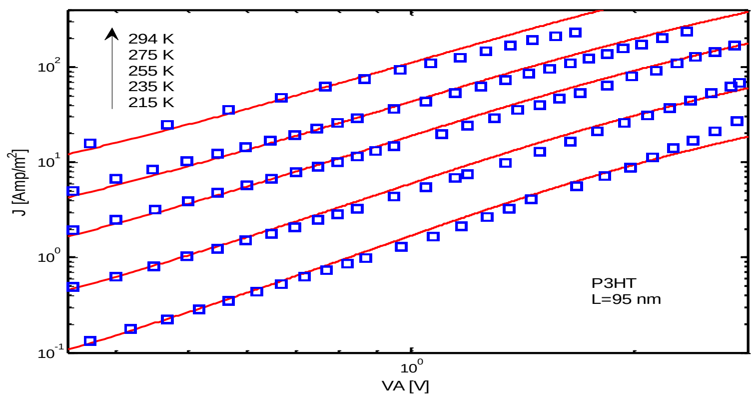

Fig.2 (color online). By applying with Single Gaussian DOS mobility edge model; (symbols represent experimental data, solid lines represent theoretical results)

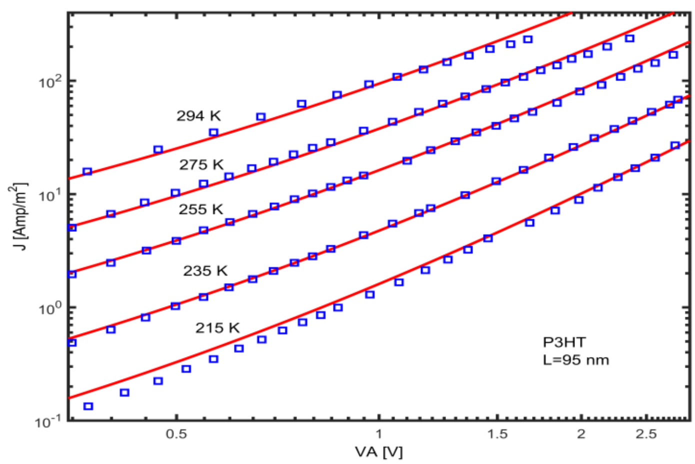

Fig.3 (color online). By applying double Gauss model; (symbols represents experimental data, solid lines represent theoretical results)

Table 1 represents the values of W_{left} and W_{right} which are independent parameters in all three models used in this literature. Table 2 represents the temperature dependent parameters used in three different models. In tables 2, we list values of N_f, μ_p and γ at every temperature for P3HT, we fit N_f, μ_p and γ by using Eqs. (11), (15) and (16) respectively. The extracted parameters for N_f, μ_p and γ of exponential model are listed in Table 3. The relationship of μ_p, and γ with temperature obey Eq. (15), and Eq. (16) respectively.

Table 4 represent the independent parameters of temperature for single Gaussian DOS mobility edge model for; including N_0, $E_{vf} = E_v - E_f$, $E_{mv} = E_m - E_v$, and σ. Value of E_{vf} is negative.

Table 1 Independent Parameters of Temperature for Exponential Model, Single Gaussian DOS Mobility edge model, and Double Gauss model for poly(3-hexylthiophene) P3HT.

	Exponential model	Mobility edge model	Double Gauss model

W_{left} (eV)	0.3	1.165	0.35
W_{right} (eV)	0.4	0.2	0.69

Table 2 Dependent parameters of Temperature for exponential model, Single Gaussian DOS mobility edge model, and Double Gauss model for poly(3-hexylthiophene) P3HT.

	T (K)	215	235	255	275	294
Exponential model	N_f (m^{-3})	7.232E2	2.1379E2	0.933E2	0.5305E2	0.368E2
	$\mu_p(0)$ (m^2/Vs)	1.0786E-	5.11E-4	1.545E-	3.061E-3	5.978E-
	γ(m/V)$^{1/2}$	0.00083	0.00069	0.00060	0.00053	0.00045
Mobility Edge model	μ_0 (m^2/Vs)	1.2E18	9.2E15	1.65E14	4.5E12	3E11
Double Gauss model	N_f (m^{-3})	8.2E32	1E32	2.12E31	4.6E30	1.5E30
	μ_0 (m^2/Vs)	22E-5	23E-5	24E-5	25E-5	26E-5

Table 3 Independent parameters of Temperature for functions $N_f(T)$, $\mu_p(0)$, and γ of exponential model for poly(3-hexylthiophene)P3HT.

N_0 (m^{-3})	E_{vF} (eV)	σ (eV)	μ_0 (m^2/Vs)	Δ_1 (eV)	Δ_2 (eV)^2	γ_0 (mV^{-1})$^{1/2}$	Δ_3 (eV)	Δ_4 (eV)^2
4.4E22	-0.3601	0.11	9.0383E-4	0.2812	-0.0059	-6.4551E-5	-0.0655	-0.0035

Table 4 Independent parameters of Temperature for Single Gaussian DOS mobility edge model for poly(3-hexylthiophene) P3HT.

N_0 (m^{-3})	E_{vf} (eV)	E_{mv} (eV)	σ (eV)
6.5E26	-0.26	0.118	0.387

Table 5 Independent parameters of Temperature of double Gaussian model for poly(3-hexylthiophene) P3HT, and for $N_f(T)$ and $\mu_0(T)$ parameters

σ_t (eV)	N_t (m^{-3})	E_{tF} (eV)	N_0 (m^{-3})	E_{vF} (eV)	σ (eV)	μ_{00} (m^2/Vs)	Δ (eV)	λ
0.15	1.7E26	0.215	3.3709E28	-0.2524	0.1305	5.755E-8	0.4744	-0.0279

The parameters in Table 5 refers to our modified model yields that σ_t ~ 0.15 eV, N_t ~ 1.7×10^{26} m^{-3}, ($E_{tF} = E_t - E_F$) ~ 0.215 eV. The μ_0 can be analyzing by using following expression with three coefficients μ_{00}, Δ and λ.

$$\mu_0 = \mu_{00} \exp\left[\Delta/kT + \lambda(\Delta/kT)^2\right] \quad (36)$$

The mobility for P3HT organic material in our modified double Gauss model is more reasonable.

Conclusion

As applying the three different mobility model to organic semiconductor polymer P3HT layer. These three models are different from one another. First the exponential model for only free carriers, the second one is the Mobility edge model for free carriers & trapped carriers, and the third model we used in this paper that is double Gauss model with our modification which consist of two Gaussian DOS (free carriers DOS, and trapped carriers DOS). The obtained results show that our modified double Gauss model is more suitable model for organic materials; it can be useful in device modeling based on organic materials. And the relationships of extracted parameters with temperature are agrees with expressions proposed in literature.

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