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Analytical Modeling of Charge transport in Bilayer Organic Light Emitting Diode

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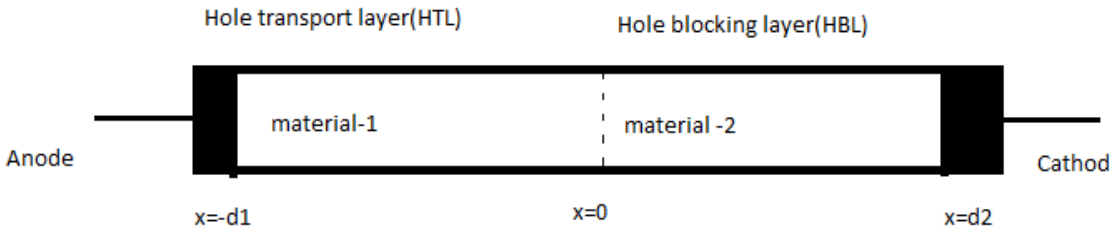
Abstract: **Charge transport, with charge carrier mobility as main parameter, is one of the fundamental properties of semiconductors. In disordered systems like most organic semiconductors, the effective mobility is a function of the electric field, the charge carrier density, and temperature. Transport is often investigated in a space-charge limited current (SCLC) regime in thin film single carrier devices, where an electric current is driven in the direction perpendicular to the surface. Direct evaluation of the current–voltage characteristics, however, is problematic, because parasitic contributions from injection or extraction barriers can falsify results.[1] Understanding and modeling charge transport in conjugated polymers is crucial to optimize the performance of organic semiconductor devices such as light-emitting diodes (OLEDs), field-effect transistors (OFETs) and solar cells[2]. The charge-carrier mobility is the major determining factor for the speed of electronic devices. Arguably, the most significant of them is the challenge to achieve a comprehensive understanding of the fundamentals of charge injection and charge transport in organics [3]. In the present work, analytical models are developed for bilayer devices in particular, which reveals the dependence of current, efficiency etc. on various device parameters.**

Keywords: **Charge transport, semiconductors, current–voltage characteristics, temperature.**

1. INTRODUCTION

Carrier injected into the organic layer can either recombine in the film or they can traverse the structure and reach the other contact without recombine carriers that traverse the device without recombining are wasted and the structure must be designed to minimize this possibility [4].since the selection of electrode material and active component on the premise of their work function as well as their oxidation and reduction potentials is limited ,one has to resort to more elaborate strategies, not only to improve injection, but at the same time to make optimum use of carriers once injected bilayer diodes employing two organic layers with different electronic energy structure and different charge carrier mobilities, allow these criteria to be more readily satisfied.[5,6]

Bilayer structure makes use of two layers of different organic material. At the interface between two organic layers there is sizeable offset in energies of the HOMO and / or LUMO level of two layers [7]. Organic material which transports hole efficiently is referred to as hole transport layer [HTL], and one which block passage of hole is referred to as hole blocking layer[HBL] as shown in figure 1 offset of the interface impedes the passage of carriers across the LED. This result in internal charge accumulation ,,redistribution of electric field ,and associated feedback effect concerning the injection of hoe and electron as well as branching between the recombination and drift towards the contact [7,8]. Thus the relative energy levels and mobilities of electrons and holes of organic materials are important device design parameters, which play an important role in the operation of the device. To provide framework for understanding bilayer device ,the effect of energy level discontinuities at organic heterojunction are first studied in a bilayer single carrier (hole only) structure .we develop analytical model that describe current density-voltage characteristics of bilayer hole-only device in the forward as well as reverse direction .these models explain the effect of hole energy level discontinuity on the current-voltage characteristics and provide insight into operation of the device.



(a) schematic diagram of a bilayer organic LED

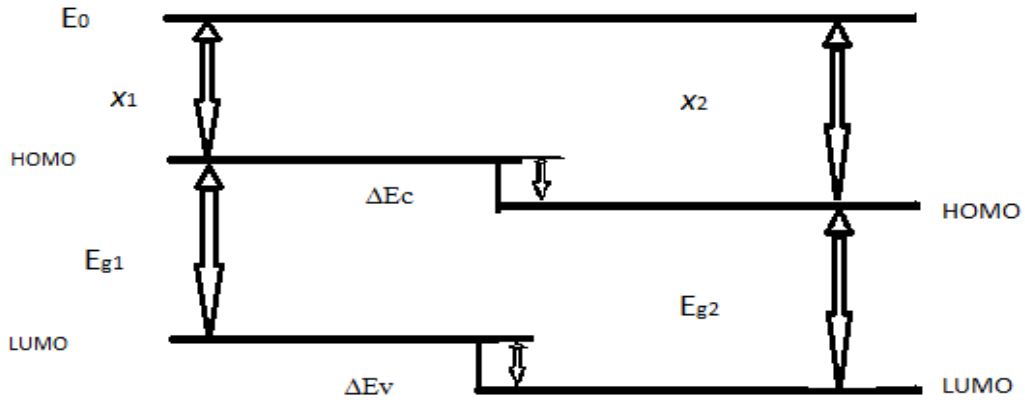


Fig 1 (a): schematic diagram of a bilayer organic LED. (b) Associated energy band diagram

2. ANALYTICAL MODEL

We consider a bilayer organic device structure with material parameters typical of the conjugate organic materials used in OLED's. The basic structure of OLED consists of metal / material-1/material-2/metal act as hole transport layer [HTL] and material-2 act as hole blocking layer [HBL]. Figure 1 shows structure of bilayer device with the associated band diagram .for sake of simplicity, we assume that the conduction energy level E_c is same for both the organic materials, and there is only a valance band discontinuity at the organic interface. The hole injection barrier at the left contact is small (0.2 eV), and electron injection barrier at the right contact is large enough to essentially make it a hole-only device.The interface is assumed to be free of sheet of charge, dipole, or recombination, so that there is continuity of electric displacement '**D**' and the electrostatic potential ø across the interface. During initial analysis, the hole mobility is assumed to be constant and same in both the materials. Both the materials are assumed to have the same dielectric constant and the transport of holes in both the materials is assumed to be trap-free. The analytical model assumes drift- diffusion transport in the bulk and thermionic emission at metal-organic interface. It is well known that under bulk –limited and trap-free condition the current in a single layer hole-only device is described by space charge limited current (SCLC) flow

$$J = \frac{9}{8}\epsilon_0\epsilon_r\mu_p(V^2/d^3) \qquad \ldots.1$$

Where the symbols have their usual meaning. The presence of 'valance-band' discontinuity of height ΔEv might be expected to reduce SCLC current in a hole-only bilayer device by a factor of exp (ΔEv /kT) so that the current density is given by

$$J = \frac{9}{8}\epsilon_0\epsilon_r\mu_p(V^2/d^3) \times exp(-\Delta E_v/kT) \qquad \ldots\ldots.2$$

In order to investigate the validity of this simple picture, numerical simulations were carried out using D D simulator [17] which is based on drift-diffusion model in the bulk and thermionic emission at the metal-organic interface Fig2 shows comparison of the result in a hole-only bi-layer device obtained with the help of numerical simulation and those predicted by Eq. (2) under forward bias condition .it can be seen that not only its magnitude of the current predicted by the sample picture an underestimation of value obtained by numerical simulation, but the slope of the curve is also larger than 2, indicating that the current density can no longer be modeled as space charge limited. Although simulation do indicate that the current scales as *~exp(ΔE/kT)*, it is clear barrier at the organic interface plays a more complex role.

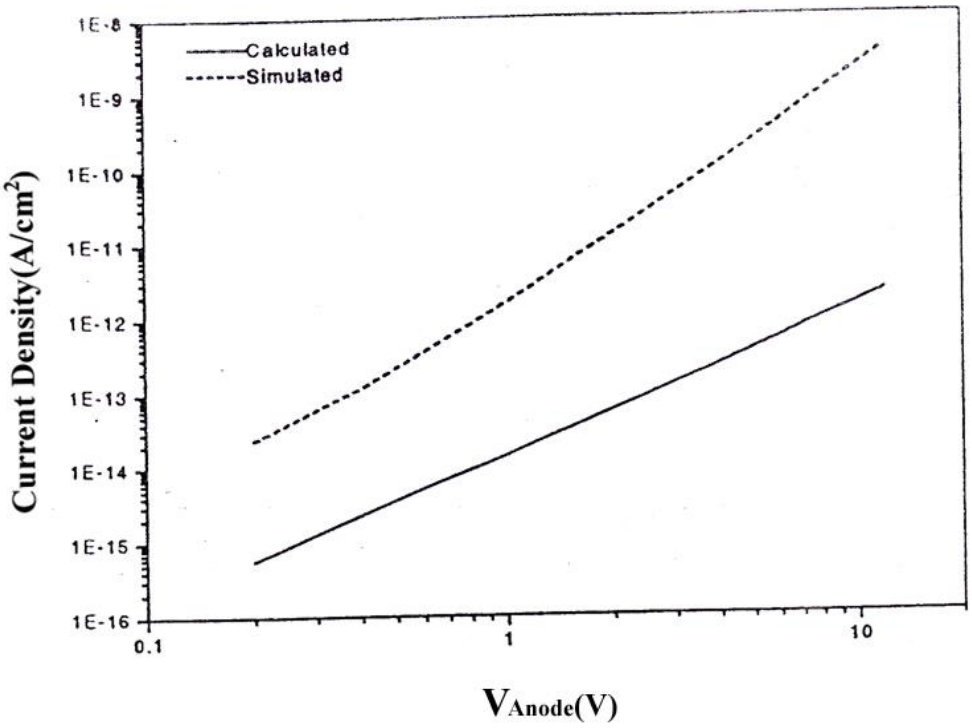


Fig 2 Current density as a function of applied bias obtained from numerical simulation (dotted line) of device shown in Fig2.1 and that obtained from Eq.2.2.2 (solid y isline).The electron affinity is 3.0 eV for both the organic material. the band gap for organic material- 1is 2.4eV and for organic material-2 is 3.0 eV the organic layer for each 500Å thick and have the same hole mobility $\mu_h$as 0.5×10-6 cm2/V-sec

Fig 3a and fig 3b shows spatial variation of the hole density and the electric field as a function of 'valance –band' discontinuity at the organic interface. the simulation results Show that for large hole barrier at the organic interface. hole density to the left of interface $_{p(0^-)}$ is much larger than the hole density to the right of interface $_{p(0^+)}$spike in the hole density to the left of organic interface causes rapid change in the electric field at the interface. Thus, field in the hole blocking layer (HBL) is of 0.6 eV at the organic interface, almost all the electric field lies in the hole blocking layer (HBL)

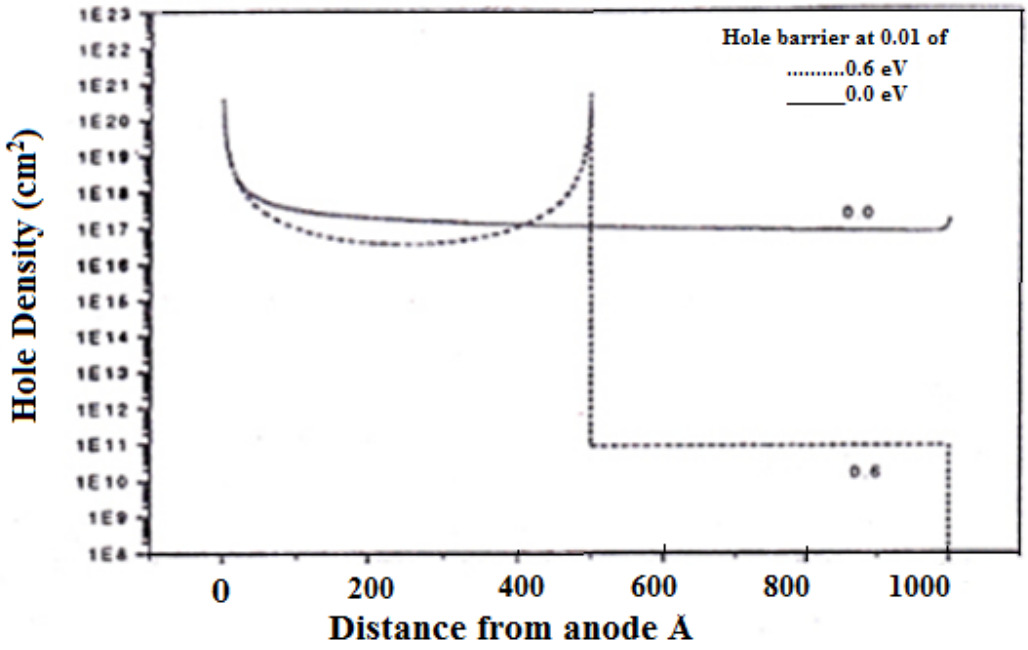


Fig 3 a: simulated variation of hole density profile for bilayer device as shown in Fig 1 as a function of hole energy level discontinuity at the organic interface

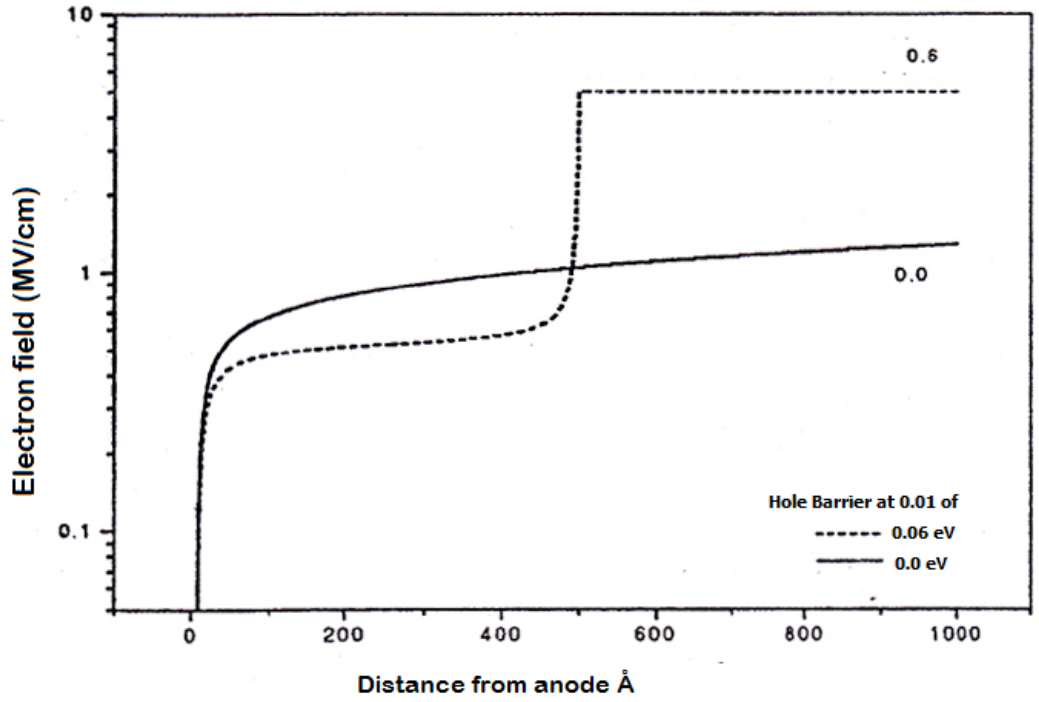


Fig 3b: simulated variation of electric field profile for the hole only as shown in fig 1 bilayer device as a function of hole energy level discontinuity at the organic interface

The electric field in the blocking layer is almost independent of the position, and increases rapidly with 'valence-band' discontinuity at the organic interface. This is in contrast to the electric field in the transport layer, where field is small and increases slowly with the 'valence-band' discontinuity. Thus addition of blocking layer has two important effects, it results in accumulation of charge in the vicinity of organic interface by impeding the flow of carriers, and because of charge accumulation there is redistribution of electric field in the structure. This accumulation of holes at the interface tends to offset the tendency of the barrier to reduce carrier injection thereby resulting in larger current values.

In the absence of any dipole layer at the interface ,the electric field would be continuous across the interface so that drift component of hole current to the left of the interface $J_p^{drift}(0^-)$ would be much larger than drift component of hole current to the right of interface $J_p^{drift}(0^+)$the only way in which continuity of hole current across the interface can then be maintained is if there is a large component of hole diffusion current to the left of the interface which is practically equal and opposite to the hole drift current.

$$J_p^{drift}(0^-) \approx J_p^{drift}(0^+) \qquad \ldots\ldots\ldots \quad 3$$

The presence of diffusion current, which opposes the drift current ,implies positive gradient in hole concentration and thus accumulation of hole at the interface Eq (.3) also implies quasi-equilibrium can be assumed to exist immediately to the left of the organic interface, an important result that would be found very useful later in the derivation of the analytical model. From fig 3b it can be assumed without much error that the entire voltage across the device is dropped across the hole blocking layer

$$E(x \geq 0) \approx (V - V_{bi})/d_2 \qquad \ldots\ldots\ldots\ldots\ldots 4$$

Where V is the applied bias and V_{bi} is the built-in voltage. With the help of this expression, hole current in the hole-blocking layer can be expressed as:

$$J_p = qp(0^+)\mu_p \times (V - V_{bi})/d_2 \qquad \ldots\ldots\ldots\ldots\ldots 5$$

The hole concentration in the vicinity of organic interface can be related to the hole barrier of height ΔE_v by:

$$p(0^+) = p(0^-) \times exp(\Delta E_v/kT) \qquad \ldots\ldots\ldots\ldots\ldots\ldots \quad 6$$

as it is clear from the simulation results that the presence of the barrier results in large accumulation of holes at the left side of interface. If we assume

$$p(x) \ll p(0^-) \ for\ x < 0^-$$

i.e. hole density elsewhere in the hole transport layer can be neglected in comparison with the hole density at the interface (Q_p) then application of Gauss's law at the interface gives

$$Q_p \approx \epsilon_0\epsilon_r \times (V - V_{bi})/d_2 \qquad \ldots\ldots\ldots\ldots\ldots\ldots\ldots \quad 7$$

The simulation results also show that the hole density decays rapidly away from the interface in hole transport layer (HTL) which we assume to be exponential in nature.

$$p(x) = p(0^-) \times exp(\delta_x)\ for(x < 0) \qquad \ldots\ldots\ldots\ldots \quad 8$$

Integration of Eq. (8) and substituting in Eq. (.7) gives

$$p(0^-) = (\epsilon_0\epsilon_r/q) \times \delta \times (V - V_{bi})/d_2 \qquad \ldots\ldots\ldots\ldots\ldots\ldots\ldots \quad 9$$

Eq. (8) together with Eq. (3) implies quasi equilibrium at the organic interface and yields an expression relating electric field at the interface and parameter δ:

$$E(0^-) = (kT/q) \times \delta \qquad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots \quad 10$$

Eq. (9) and Eq.(10) along with Eq.(.4) provides means of relating $p(0^-)$ with the applied voltage as:

$$p(0^-) = (\epsilon_0\epsilon_r/kT) \times [(V - V_{bi})/d_2]^2 \qquad \ldots\ldots\ldots\ldots\ldots\ldots \quad 11$$

Substituting Eq. (11) and Eq. (6) in Eq. (5) we arrive at the central expression for current density:

$$J_p = \epsilon_0\epsilon_r \times \mu_p \times q/kT \times [(V - V_{bi})/d_2]^3 \times exp(\Delta E_v/kT) \qquad \ldots\ldots\ldots\ldots \quad 12$$

But Eq.(12) is valid only for interface barriers, which are large enough to justify the assumptions made derivation .As the barrier reduces in the limit $\Delta E_v \rightarrow 0$ the current predicted by Eq (12) does not to go to the SCLC limit. This drawback can be rectified by casting Eq. (12) In a more instructive form as:

$$J_p = \left(\frac{9}{8}\right)\epsilon_0\epsilon_r\mu_\rho\left[\frac{(V - V_{bi})^2}{d^3}\right] \times \exp(-E_V/kT) \times \left[\left(\frac{8}{9}\right) \times \left(\frac{d}{d_2}\right)^3 \times \left(\frac{(V - V_{bi})}{kT/q}\right)\right]$$

…. .13

While the first term describes the conventional space charge limited current, the second term describes the role of the barrier in reducing carrier injection into the hole - blocking layer. The last term can be looked upon as the effect of barrier in causing accumulation of the interface. A direct consequence of Eq. (13) is new expression, which would correctly predict the behavior from large interface barrier down to zero interface barriers as well:

$$J_p = \frac{\frac{9}{8} \times \epsilon_0\epsilon_r\mu_p \times \frac{(V - V_{bi})^2}{d^3}}{[1 + exp(\Delta E_v/kT)]/\left[\frac{8}{9} \times \left(\frac{d}{d_2}\right)^3 \times \left(\frac{V - V_{bi}}{kT/q}\right)\right]} \qquad .14$$

3. RESULT AND DISCUSSION

Fig 4 shows that the current values predicted by the above expression matches very well with the simulation results for the barrier heights varying from 0.6eV down to 0.1eV .figure shows variation of current density as the cube of applied voltage *~$(V\text{-}V_{bi})^3$, and inverse cube of the blocking* layer thickness i.e. *~ $1/d_2^3$* eq. 14 reduces to eq..12 for the interface barrier that are larger than about 0.25eV .in this case the current is predominantly interface barrier limited and dependence of the current on both the voltage and thickness is identical however the proposed model slightly overestimates the current .this is because of two assumption made during the derivation; first it was assumed that the entire voltage drops across the hole-blocking layer, which results in overestimation of field in this region

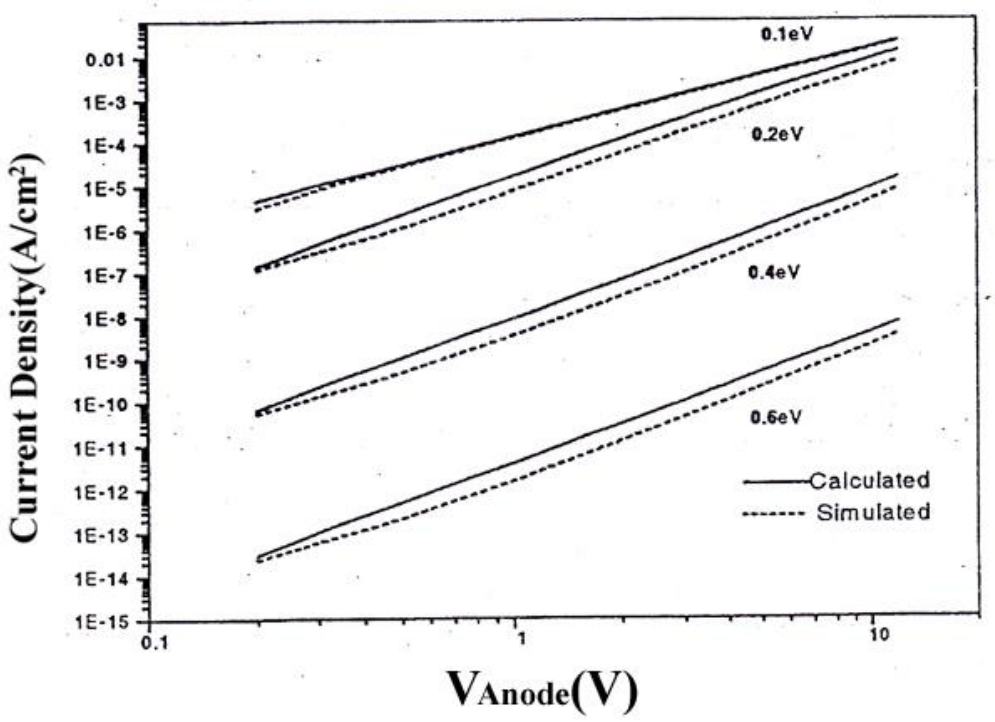


Fig 4: current density as a function of applied bias obtained from numerical simulation and obtained from eq. 14(solid line) for the hole only bilayer device as a function of different values of hole barrier height

secondly, it was assumed that hole density elsewhere in HTL, is small compared to that at the interface, which results in overestimation of hole density at the interface. For large barrier heights, it is easy to incorporate the dependence of mobility on the electric field since in the hole-blocking layer is large and uniform. Assuming $\mu_{p2} = \mu_{02} \times exp\left(\sqrt{E/E_0}\right)$, we obtain a more general expression for current density in a hole-only bilayer device for large hole interface barrier height as :

$$J_p = \epsilon_0 \epsilon_r \mu_{02} \times \frac{q}{kT} \times \left(\frac{V-V_{bi}}{d_2}\right)^3 \times exp\left(\sqrt{\frac{(V-V_{bi})/d_2}{E_0}}\right) \times exp(-\Delta E_v/kT) \qquad \ldots. 15$$

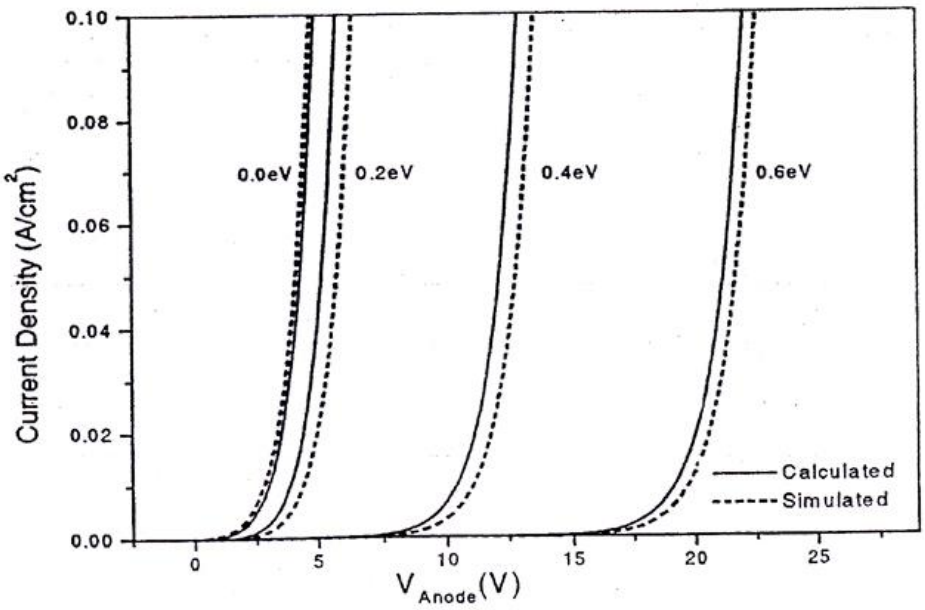


Fig 5: Current-Voltage (J-V) characteristics for hole-only bilayer device as a function of valance band discontinuity, taking into account field dependence of hole mobility.curve show increase in the voltage to attain the same value of current density of 0.1A/cm^2.

Fig.5 shows that successively higher voltages are required to obtain the same value of current density as the energy barrier to hole injection from material-1 to material-2 is increased. Even the modest 0.2eV barrier causes as increase in the voltage required to obtain a given current. The curves are obtained by taking field dependence of mobility on the basis of Eq. (15). Similar results are reported by B.K. Crone *et al. al [5]*, but with E_0 equal to $2x10^4 V/cm$. At the larger values of hole barrier at the organic interface, current at a particular voltage is significantly reduced. It is evident that even a small increase in the barrier at the organic interface can significantly increase the biasing requirements. This increase in the voltage caused by the increase of barrier at organic-organic. Even the modest 0.2eV barrier causes as increase in the voltage required to obtain a given current. The curves are obtained by taking field dependence of mobility on the basis of Eq. (15).

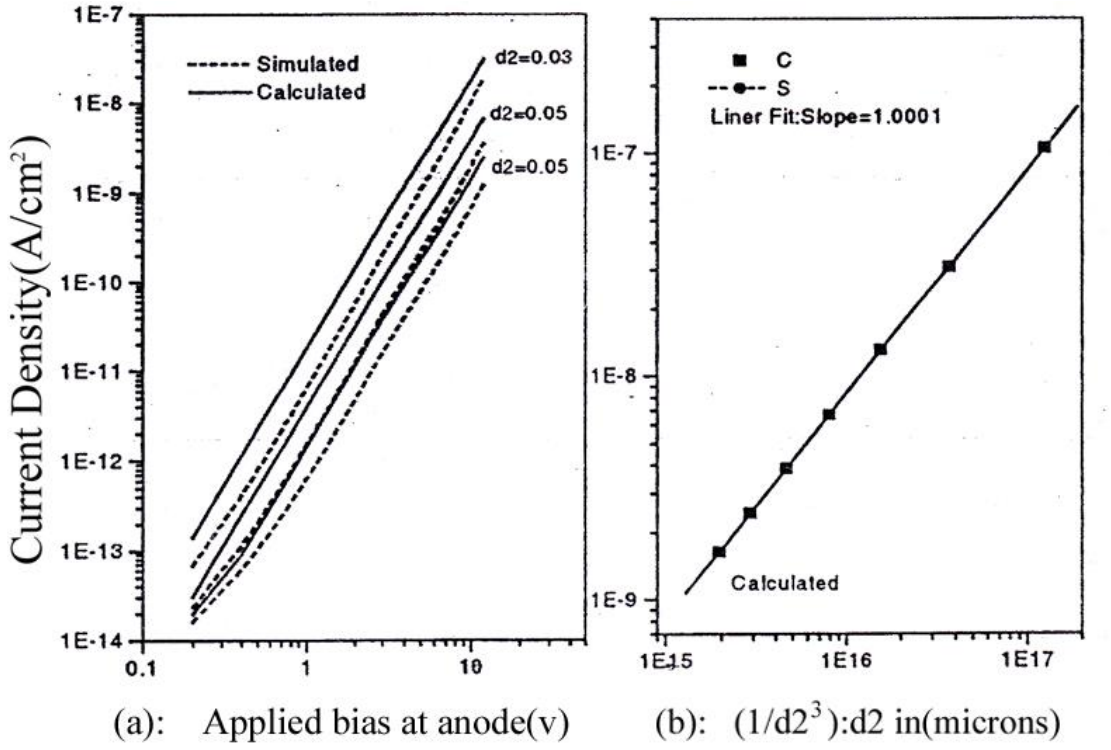


Fig 6a: Calculated and simulated current density-Voltage characteristics for a bilayer hole only device with different blocking layer thicknesses.fig 6 b variations in current density w.r.t. variation in inverse cube of blocking layer thickness

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The whole derivation was made under the assumption, that both the materials are same i.e. have same hole and electron mobility. Difference in the hole mobility also results in the accumulation of holes at the organic interface, and the impact of hole mobility discontinuity is similar to that of valence-band discontinuity. However an analysis of the steps involved in the derivation of analytical expression show that as long as there is a large accumulation of holes at the interface, the hole mobility deference in the two layers will not matter. Thus Eq. (12) will continue to hold good as long as barrier at the organic interface is large, except that μ_p has to be interpreted as μ_{p2}: mobility of the hole blocking layer. But as $\sim \Delta E_v$ 0,

Eq. (14) becomes equal to Eq. (.1) for SCLC in single layer single carrier device and the mobility should be interpreted as μ_{p1}. Though it is difficult to develop a general analytical model that is valid for arbitrary values of hole barrier heights and mobility ratios, but Eq. (14) can be modified to take into account different hole mobility values in the two organic layers both at large and small barrier height at the organic as well :

$$J_p = \frac{\frac{9}{8}\times\epsilon_0\epsilon_r\times\mu_{pt}\frac{(V-V_{bi})^2}{d^3}}{\{1+\frac{\mu_{p1}}{\mu_{p2}}\times exp(\Delta E_v/kT)\}\Big/\frac{8}{9}\times\left(\frac{d}{d_2}\right)^3\times\left(\frac{(V-V_{bi})}{kT/q}\right)} \qquad \text{.....16}$$

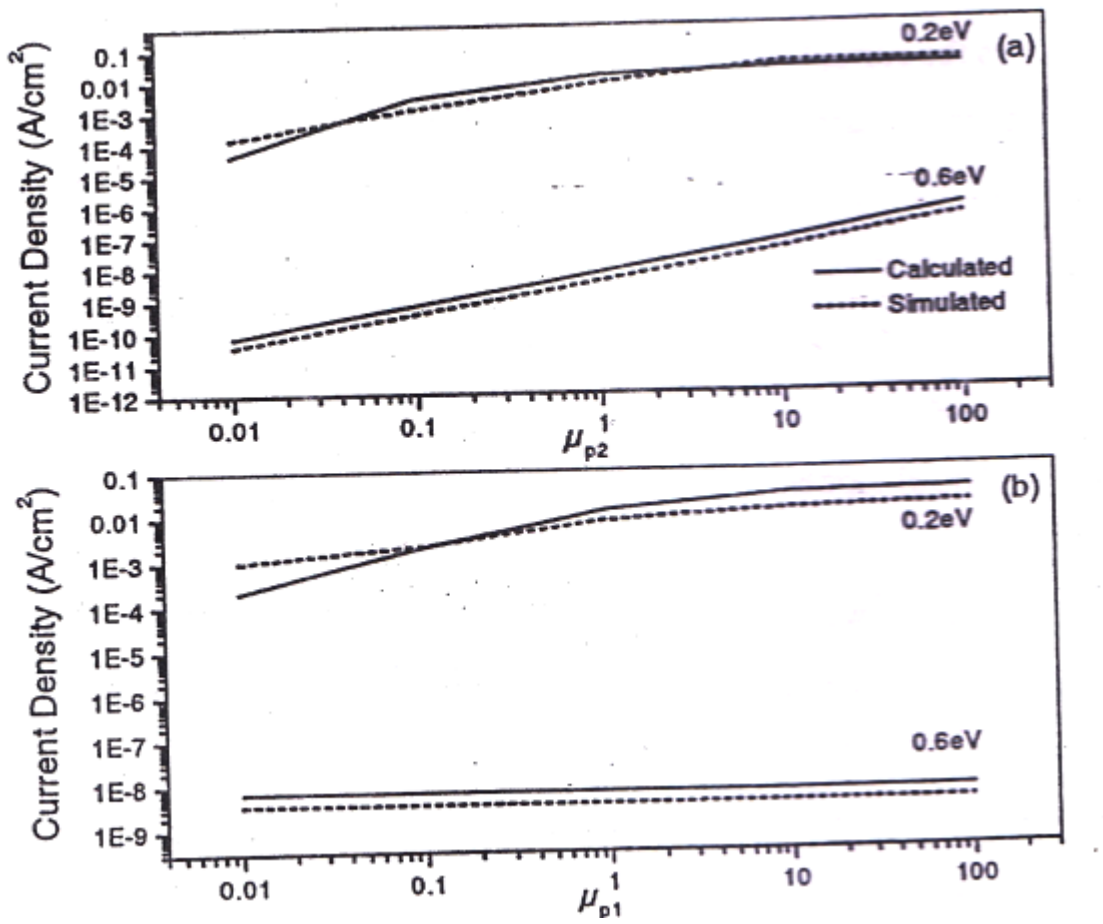

Fig 7 (a): Shows variation in the calculated and simulated current density as a function of the transport and blocking layer hole mobility ratio Fig 7 (b) transporting layer mobility μ_{p1} is varied.

Fig 7 shows the dependence of current density on the mobility ratio for both large and small barrier heights. Fig 7 a shown the current density plotted w.r.t. variation in mobility ratio, when μ_{p1} (transport layer mobility) is kept constant and mobility of blocking layer, μ_{p1} is varied. While Fig. 7b shows variation of current density w.r.t. variation in mobility ratio, when μ_{p1} is varied and μ_{p1} is kept constant. Both the figures along with equation established the face that the current density is rather insensitive to the transport layer mobility, and scale directly with the blocking layer mobility as long as barrier at the interface is kept large enough to block the transport of holes.

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