國立交通大學

物理研究所

碩士論文

載子陷阱在高分子發光二極體內的效應

Effects of Carrier Trap in Polymer Light Emitting Diodes

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博碩士論文授權書

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論文名稱：Effects of Carrier Trap in Polymer Light Emitting Diodes

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論文名稱： Effects of Carrier Trap in Polymer Light Emitting Diodes

指導教授：孟心飛，洪勝富

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Effects of Carrier Trap in Polymer Light Emitting Diodes

研 究 生：蔡明燃　　Student：Min-Ran Tasi

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國立交通大學

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Abstract

This thesis represents the effects of traps in polymer light-emitting diodes. In electron only devices,the electron conduction in polymer is strongly reduced by the presence of traps. Putting more traps, we find stronger electric field dependence in electron current density. If we apply larger voltage, because more traps are filled, electron current is approaching to the hole current density. Traps not only reduce the current in electron only devices, but also increase the time required for device reaching the steady state. In our simulation, we represent that traps contribute to the time delay in devices.

In double carrier devices, because of Coulomb force, putting more traps contribute to the injection of hole current density and total current density. In our simulation, two times total current density is possible. We consider the recombination process as bimolecular and using Langevin form for recombination coefficient. Because of the imbalance between electron and hole current density, putting more traps result in lower efficiency in polymer light-emitting diodes. If we put traps more than $10^{19}\mathrm{cm}^{-3}$, the recombination efficiency can be suppressed to one-half. We observe that when the trap density is one-tenth of the free electron density without traps, the efficiency begins to go down greatly. Similarly, we consider the time scale in bipolar devices.

Contents

1 Introduction

Polymer light-emitting diode (PLED) has been focused at since 1990 [1] because of its easy processing and mechanical flexibility. PLED consists of a thin layer(or multilayer) of conjugated luminescent organic material sandwiched between two electrodes. Because of the large band gap in organic material(>2eV), carriers in organic material are mainly injected from the electrode. So, different kinds of electrodes determine whether the device is electron only or hole only or bipolar device. Carriers can move across the device and recombine, perhaps emitting light. Up to now, there are lots of groups considering the features of PLED including electric and optical properties of this material. One of the most important characteristics of this material is the imbalance between electron and hole mobility [2]. In time-of-flight measurement shows that electrons, in contrast to holes, are severely trapped in polymer material [2]. The higher hole mobility is explained by two reasons. The first is that the hole trap caused by structure defect as electron trap are compensated by background p-doping [3]. The second is that oxidation contribute to electron trap, but not hole trap [3]. These two reasons explain why hole mobility is in general several orders of magnitude larger than electron mobility. Because of electron trapping, electron mobility is reduced, and perhaps the electroluminescence (EL)is confined close to the cathode. Since metallic electrodes are efficient quenching centers for EL, this confinement is expected to strongly reduce the device performance.

Carrier mobility in organic material also depends on the electric field [17, 4] and the field-dependence of electron mobility is larger than hole mobility. This phenomenon is explained by the appearance of electron trap, and we will show this in our calculated results later. Review of literature so far no trap with diffusion current has been considered, we consider the trap effect with diffusion current in our model.

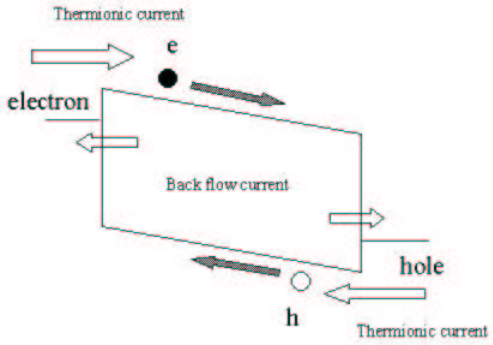
In this thesis, we focus on the effects of device with traps. We use the model used in Crone [5] and Davids [6] and consider the exponential trap distribution in energy. In electron only devices, we show that the more electron trap density, the more field-dependence in electron mobility. Electron distribution is important in devices because it determine the recombination region. In this section, we compare the distribution calculated by mobility model used in Poole-Frankel form with that determined by our trap model to understand whether the quenching effect is important or not.

In bipolar devices, we concentrate on influence of trap on the recombination rate, recombination efficiency and $J_t - V$, $J_t - N_t$ curves. We also show that the total current density is correlative to the trap density, also does the recombination efficiency. Besides, we consider the time scale that device reach the steady state. In our simulation, we observe traps will also contribute to the time delay in organic devices. For hole only devices, the device time scale is micron second(μs), but in electron or bipolar devices, millisecond (ms) time scale is possible.

This thesis is organized as follows:Sec 2 discuss the device model, Sec 3 represents and discuss the results, Sec 4 makes a conclusion, and figures are in Sec 5. Sec 6 is appendix.

2 Model

PLEDs consist of a layer(or multilayer) of organic material sandwiched between two electrodes. Carriers are injected from electrodes, and transport through the organic material. The transport of charge through the material depends on the bulk properties of the organic material and is determined by the solution of Poisson's equations coupled with continuity equations, using the drift and diffusion current density and the field dependent mobility. The scheme of injection and transport is below:



A polymer chain is consist of a series of conjugated chain segments which are terminated by twists or other defects, and that is the origin of trap. The organic materials have large band gap which result in low carrier density in thermal equilibrium. Carriers are mainly injected from electrodes. Electron and hole carriers are moving through the material by moving through the chain segments and then hopping to the next chain segment [4]. The carrier transport is limited by the hopping rate, which leads to the field dependent mobility in organic materials which have the form:$\mu = \mu_0 e^{\sqrt{E/E_0}}$. We will interpret this form later.

In our simulation, non-degenerate case are considered, therefore, the equilibrium carrier densities of hole and electron are given as :

$$n_f(x) = n_0 e^{-(\frac{\varepsilon_c - e\phi - \mu}{kT})} \tag{1}$$

and

$$p(x) = n_0 e^{(\frac{\varepsilon_v - e\phi - \mu}{kT})} \tag{2}$$

where n_f and p are free electron and hole carrier densities, n_0 is the total density of states of carriers, and μ is the chemical potential of the device. ϕ is the potential where the zero point is in the left electrode. T is the temperature of the environment, k is the Boltzman's constant. These levels ε_v and ε_c are effective conduction band and valance band such that the above expression gives the same results as:

$$n_f(x) = n_0 \int D_c(E) f dE \tag{3}$$

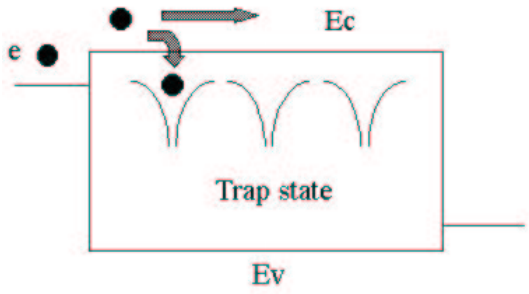
and

$$P_(x) = n_0 \int D_v(E) f dE \tag{4}$$

where D_c and D_v are the density of states of conduction and valance bands, f is Fermi-Dirac distribution. In this thesis, we consider the trap density with an exponential distribution:

$$n_{t\varepsilon}(\varepsilon, x) = (\frac{N_t(x)}{kT_t}) e^{(\frac{\varepsilon - \varepsilon_c}{kT_t})} \tag{5}$$

where $n_{t\varepsilon}(\varepsilon, x)$ is the trap density of states at energy ε, ε_c is the conduction band energy. $N_t(x)$ is the total trap density of states, and kT_t is an characteristic energy of trap distribution. Because of the p-doping in our material, the hole traps are considered as totally full-filled, so that we don't consider the hole trap in our model. We only consider the electron trap in our simulation [3].



The transport property is treated by solving the continuity equations coupled with Poisson's equation and drift-diffusion current:

$$\frac{\partial n_{t0}}{\partial t} = \frac{1}{e}(\frac{\partial J_n}{\partial x}) - R \tag{6}$$

$$\frac{\partial p}{\partial t} = -\frac{1}{e}(\frac{\partial J_p}{\partial x}) - R \tag{7}$$

where

$$J_n = e\mu(nE + \frac{kT}{e}\frac{\partial n_f}{\partial x}) \tag{8}$$

$$J_p = e\mu(pE - \frac{kT}{e}\frac{\partial p}{\partial x}). \tag{9}$$

n_{t0} is the total electron density, including the free and trapped electrons, J_n and J_p are free electron and hole current density. E is the electric field, and R is the recombination rate per unit volume, e is the absolute value of electron charge, and μ is the carrier mobility. The carrier hopping property is characterized by the field dependent mobility ,

seen in time-of-flight measurement [11, 12, 13, 14]. For devices at given temperature, the mobility is given by [4, 10]:

$$\mu = \mu_0 e^{\sqrt{\frac{E}{E_0}}} \tag{10}$$

and

$$\mu_0 = \mu^* \ e^{-(\frac{2}{3}\beta\sigma)^2} \tag{11}$$

where β is 1/kT. The form of mobility is proposed by Gill and later interpreted by others in terms of the dipole trap model where the transport sites are dipolar trap in polymer matrix [17, 18]. The E and T dependence arise from the variation of the escape rate from the dipolar traps. σ is the deviation of the Gaussian distribution of site energies, and μ^* is intrinsic mobility of carrier in organic material which is independent of T and E. Equation (10) based on the gaussian disorder model of Bässler, where carrier is hopping through the sites with Gaussian distributed energies. The mobility with field and temperature dependence is the so called Poole-Frenkel (PF)mobility. In PLED, the μ_0 of electron is smaller than that of hole in several orders of magnitude, and the E_0 of electron is smaller than that of hole. These effects that electron and hole mobility are different result from the appearance of traps, and we call the model that electron and hole have different mobility Poole-Frenkel (PF)model. In this thesis, we directly consider the trap effect in which the free electron and hole mobilities are the same. Meanwhile, electrons could be trapped by traps, and we could see this effect that electron current is reduced by the appearance of traps. We call our model trap model.

In our trap model, we do not consider the generation rate for the fact that the band gap of this material is too large ($\geq$ 2eV)to generate enough carriers compared with that from injection. We model the recombination process as bimolecular with the form[6, 7, 11]:

$$R = r n_f p \tag{12}$$

where n_f and p are free electron and hole density, and r is a coefficient. We also use the Langenvin form for r, and this form assumes the recombination is rapid when electrons and holes are moving toward to each other rapidly. The coefficient can be written as:

$$r = \frac{4\pi e \mu_r}{\epsilon_0 \epsilon_r} \tag{13}$$

where μ_r is effective mobility which is taken to be the larger one of the electron and hole mobility in PF model[12]. In our trap model, we make effective mobility as carrier mobility. ϵ_r is the relative permittivity of organic material.

In our works, we solve the equations numerically using Sharfetter-Gummel special discretization and integrating the continuity equations from equilibrium state to steady state, which is the so called Gear's method. For example:

$$\frac{p(t+\Delta t) - p(t)}{\Delta t} = \frac{-1}{e} \frac{J_p(x+\Delta x) - J_p(x)}{\Delta x} - r n_f(x) p(x) \tag{14}$$

$$\Rightarrow p(t+\Delta t) = p(t) - \frac{\Delta t}{e}\frac{J_p(x+\Delta x) - J_p(x)}{\Delta x} - \Delta t r n_f(x) p(x).$$

In this way, we can find the the carrier density in next time. In order to use the Gear's method, thermal equilibrium solution E, p, n_f and n_t is necessary. We solve the Poisson's equation to find the E, p, n_f and n_t in thermal equilibrium , that is:

$$\frac{\partial E}{\partial x} = \frac{e(p - n_f - n_t)}{\epsilon} \tag{15}$$

and

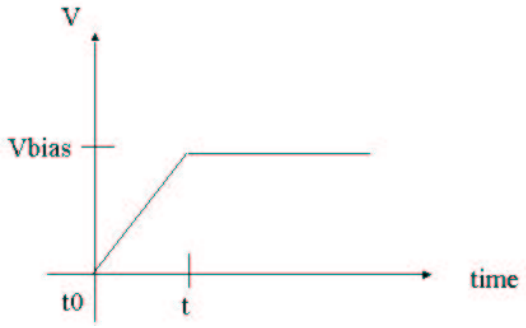
$$\frac{\partial \phi}{\partial x} = -E. \tag{16}$$

We use equations (1),(2), (15), (16), (49) and the relaxation method to find out the solutions. Besides, the boundary condition is the work functions of two electrodes.

Using time derivative to Poisson's equation, we can find the time derivative of electric field [6]:

$$\frac{\partial E(x,t)}{\partial t} = \frac{-1}{L}\frac{\partial V_L}{\partial t} - \frac{1}{\epsilon_0 \epsilon_r}[J_t(x) - \frac{1}{L}\int_0^L J_t(x)dx] \tag{17}$$

where V_L is the potential at right contact and J_t is the total current at x, L is the thickness of the device. We derive this equation in Appendix A. Here, voltage ramp from t_0 to t is applied, starting from the equilibrium solution already obtained, and preceding stepwise through bias of interest.



When the total bias is applied at time t, the first term in the right hand side of equation (17) is vanished after time t, and the system begin to reach the steady state by means of the second term. In steady state, the total current density $J_t = J_n + J_p$ is constant respected to x, and we use this as our condition of system reaching steady state.

We also want to find out the time derivative of n_t and n_f so as to use the Gear's method to find out the solution in steady state. n_t is the trapped electron density. Because the trapped electrons are the electrons which are scattered by phonons, and this process ($\sim 10^{-12}$ sec)is faster than the time scale in which device reach the steady state ($\sim 10^{-6} sec$) [16], we assume that free electrons and trapped electrons are locally quasi-thermal equilibrium [11, 14]. We also assume that Fermi-Dirac distribution is a step

function because the characteristic energy of trap distribution $kT_t(\sim 0.15\text{eV})$ is larger than thermal energy kT in room temperature($\sim 0.0259\text{eV}$) [15]. Using these assumptions, we observe the time derivative of n_f and n_t are:

$$\frac{\partial n_f}{\partial t} = \frac{1}{1+B}[\frac{1}{e}\frac{\partial J_n}{\partial x} - rn_f p] \tag{18}$$

$$\frac{\partial n_t}{\partial t} = \frac{B}{1+B}[\frac{1}{e}\frac{\partial J_n}{\partial x} - rn_f p] \tag{19}$$

where

$$B = Nt\frac{T}{T_t}(\frac{1}{n_0})^{T/T_t}\frac{1}{n_f^{1-T/T_t}}. \tag{20}$$

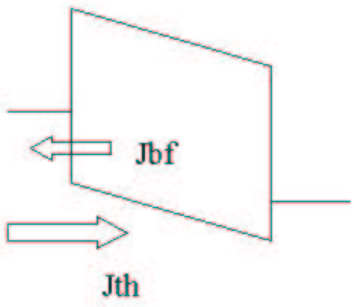
We derive equations (18)(19)(20) in Appendix B. From now on, we can use Gear's method to find out the steady state solutions:n_f, n_t, p and electric field E.

Carrier injection provides a boundary condition for solving the equations. In contacts we consider the thermionic emission and back flow current which is the time reversed process of thermionic emission. These currents add up to total current density. For instance, the total hole current density at left contact is :

$$J = J_{th} - J_{bf} \tag{21}$$

where

$$J_{th} = AT^2 e^{-\frac{\phi_b}{kT}}. \tag{22}$$



ϕ_b is the Schottky barrier high, A is the Richardson constant. Back flow current is considered as proportional to the carrier density near the electrode. If we consider the hole back flow current at left contact, that is:

$$J_{bf} = \nu p(x=0). \tag{23}$$

In equilibrium, thermionic and back flow current must cancel out, we have:

$$AT^2 e^{-\frac{\phi_b}{kT}} = \nu p(0) \tag{24}$$

and we use the hole carrier density in equilibrium:

$$p(x=0) = n_0 e^{(\frac{\varepsilon_v - e\phi - \mu}{kT})} = n_0 e^{-\frac{\phi_b}{kT}} \tag{25}$$

$$\Rightarrow \nu = \frac{AT^2}{n_0}. \tag{26}$$

So the total hole current density at left contact is given by :

$$J_p(0) = \frac{AT^2}{n_0}(n_0 e^{-\frac{\phi_b}{kT}} - p(0)) \tag{27}$$

$$= \frac{AT^2}{n_0}(p_{eq}(0) - p(0)) \tag{28}$$

where the p_{eq} is thermal equilibrium hole density in the left contact. We also consider the image force lowing for ϕ_b if the field has the correct sign, that is [5, 6]:

$$\phi_b = \phi_{b0} - e\sqrt{\frac{e\|E\|}{4\pi\epsilon_0\epsilon_r}} \tag{29}$$

where $\|E\|$ is the absolute value of electric field at contact. In steady state, we can integrate the continuity equation (6), (7) to obtain the recombination current J_r:

$$J_r = \int_0^L eRdx = J_n(L) - J_n(0) = J_p(0) - J_p(L). \tag{30}$$

When electrons(or holes) move cross the device, it is possible to recombine with holes(or electrons), and this is the origin of J_r. Both electron current at the hole injection contact and the hole current at injection electron result in the loss of recombination efficiency. The efficiency that charge carrier transfer to photon is given by:

$$E_{ff} = \frac{J_r}{J_t} \tag{31}$$

where J_t is total current density.

3 Results and Discussion

3.1 Electron Only Devices

In this subsection, we consider the electron only devices with traps. In polymer light emitting diodes, electrons are trapped and the electron mobility is smaller than that of hole for 2 or 3 orders of magnitude. We consider organic diodes fabricated using MEH-PPV with electron and hole energy levels to be $\varepsilon_c = 2.9$eV and $\varepsilon_v = 5.3$eV. In our model , we consider devices with cathode in left hand side and anode in right hand side. We also assume dielectric constant $\epsilon_r = 3$ and $n_0 = 10^{21}$ cm^{-3}. To begin with, we consider the electron only device with two calcium(Ca) electrodes and electrons are injected from which to MEH-PPV with 0.2 eV barrier high.

The first of all, we want to compare the results calculated by trap model and PF model, especially in electron density distribution. In Fig-1, we use our trap model to plot the $J_e - V$ curve with $N_t = 10^{18}$ cm^{-3}, $L = 100nm$, $T = 300$ $Kelvin(K)$, and characteristic temperature of trap $Tt = 1500$ K. We assume $\mu_0 = 10^{-6}$cm^2/Vs and $E_0 = 10^5$V/cm, $n_0 = 10^{21}$ cm^{-3}. We fit this figure using Poole-Frenkel (PF) model in Fig-2 with parameters μ_0 and E_0 of electron mobility are 8×10^{-11} cm^2/Vs and 1.9×10^4 V/cm, other parameters are the same as before. We have almost the same curves in trap and PF model. Now, we compare their results in electron distribution in Fig. 3 and 4. When we apply larger voltage, electron density distribution become larger in both cases. In PF model, because the electron mobility is smaller than that in trap model, the electron distribution is larger than that in trap model to create the same I-V curve, especially near the cathode. This result in the importance of quenching effect because the electron density near the cathode is very large($\sim 10^{19}$ cm^{-3}). In trap model, although there are traps through device, the free electron mobility is the same as hole's, and that is why electrons could have the same I-V curve with smaller carrier density. Quenching effect in devices is important, and in this case we observe that if we want to consider the quenching effect, the trap model is better than PF model. Fig. 5 shows that the traps in device result in the lowing of electron current density. In Fig. 5, $n_0 = 10^{21}$ cm^{-3}, $\mu_0 = 10^{-6}$, and $E_0 = 5 \times 10^5$V/cm, T is 300 K and T_t is 1500 K. The thickness of device L is 100 nm. We observe that there are critical total trap density in which the current lowing become apparent. Because the free electrons and trapped electrons are under thermal equilibrium, so that if we introduce traps, the more trapped electrons appear in device with more free electrons (that is, electrons trapped in 8 volt is more than that in 2 volt). That is why the critical total trap density of device with larger voltage is a little smaller than that in smaller voltage.

In Fig. 6, we concentrate on the time scale in which device reaches the steady state. $n_0 = 10^{21}$ cm^{-3}, $\mu_0 = 10^{-6}$, and $E_0 = 5 \times 10^5$V/cm, T is 300 K and T_t is 1500 K. The thickness of device L is 100 nm. We can observe that there are also critical total trap densities. In Fig. 6, for smaller voltage, device needs more time to reach the steady state. It is reasonable because the phonon scattering is very fast, so that electrons must fill up traps and then go through the device. Smaller voltage results in not so many electrons, and this is why the filling time is longer. This effect is also the origin of critical trap

density in Fig. 6. When the trap density is dense enough, electrons begin to spend much time filling up the traps. We find when the trap density is one-tenth of the averaged free electron density of device without trap, rising of time scale is apparent. That is:

$$n_t = \frac{n_f}{10} = N_t(\frac{n_f}{n_0})^{T/T_t}. \tag{32}$$

Note that the result obtained in Fig. 6 is in contrast with that in Fig. 5. In Fig. 5, larger voltage will have smaller critical trap density, and in Fig. 6, the result is reversed.

In PLED, the electron mobility has more field dependence observed in time-of-light experiment (that is, E_0 of electron is smaller than hole's) [2], and we want to observe this effect in our trap model. Fig. 7 shows that field dependence of electron current is correlative to the trap density in devices. Here $n_0 = 10^{21}$ cm^{-3}, $\mu_0 = 10^{17}$cm^2/Vs, $E_0 = 10^5$V/cm, L=100 nm, T and T_t are 300 and 1500 K. We set mobility being constant (that is, $\mu = \mu_0$)because the more field dependence of electron mobility result from the appearance of traps, but not from the intrinsic transport property in organic material. In this figure, we also plot the space-charge limited current(SCLC) in which [9]:

$$J = \frac{9}{8}\epsilon_0\epsilon_r\mu\frac{V^2}{L^3} \tag{33}$$

where μ is the carrier mobility, V is the applied voltage. The SCLC curve shows the carrier current density of device without trap in bulk-limited regime. When we put rare trap densities N_t, it is reasonable that the electron current is just the same as current without trap. Applying larger voltage, the traps are filled, and the current should be approaching to the SCLC. We observe our current density approaching to SCLC when we lowing the trap density or applying larger voltage. The current is almost the same with trap density below 10^{17}cm^{-3} in our condition. Equation (33) is derived in Appendix C.

3.2 Bipolar Devices

In bipolar devices, we consider the Ca/MEH-PPV/Au devices with the barrier high for hole injection from Au to MEH-PPV is 0.2eV. The characteristic temperature of trap distribution is 1500 K, and we also assume the total carrier density n_0 is 10^{21} cm^{-3}, $\epsilon_r = 3$, $\mu_0 = 10^{-6}$ cm^2/Vs, $T = 300$ K, and $E_0 = 10^5$ V/cm.

In Fig. 8, the curve $J_t - N_t$ is represented with the trap densities $1, 10^{15}, 10^{16}$, $10^{17}, 10^{18}, 2 \times 10^{18}$ and 5×10^{18} cm^{-3}. This figure shows that the total current density, in bipolar devices, is increasing with total trap density N_t increased. It is in contrast with electron only devices. In electron only devices, because some electrons are trapped, the electron current decrease when trap density N_t is increased. In bipolar devices, however, electrons are trapped which result in negative space charge in device, and finally contribute to hole injection. In Fig. 9, the hole current density is represented and current near anode (right hand side) is increasing with increasd trap density, and that is the

reason why total current density J_t increase when N_t is increased. Fig. 10 shows the possibility of two times total current density J_t when N_t is large as 10^{19} cm^{-3} in this condition.

Fig. 11 displays the efficiency of bipolar devices with parameters are the same as before. It shows when trap density become larger, the efficiency is decreasing. In devices with traps, because of the stronger imbalance between electron and hole carrier density, larger trap density result in smaller efficiency. This figure also shows the phenomenon that the smaller applied voltage also result in stronger imbalance between electron and hole carrier density. In smaller voltage, electron current is smaller than hole current(Fig. 5), and that is why the efficiency lowing with smaller voltage is faster than that with larger applied voltage.

Distribution of light generation $r \cdot n(x) \cdot p(x)(equqtion\ (12))$ is important in bipolar devices because the dominant light generation zone determine whether the quenching effect is important or not. Fig. 12 represents the feature that increase trap density, the dominant recombination is approaching to the cathode. The voltage of this device is 8volt, μ_0 is 10^{-6} cm^2/Vs, E_0 is 10^5 V/cm,$n_0 = 10^{21}$ cm^{-3}, and the thickness L is 100 nm. T and T_t are 300 and 1500 K. This is a very important feature because the quenching effect will result in the exciton energy transfer, and the exciton will not emit light. This effect will reduce the performance of device because the major recombination process happens near the cathode. Another feature in Fig. 12 is that the maximum of distribution become larger when we put more traps. We explain this phenomenon by displaying carrier distribution with different trap density in Fig. 13. We choose $N_t = 10^{15}$ cm^{-3} and $N_t = 10^{18}$ cm^{-3} in our discussion. When N_t is 10^{15}cm^{-3}, the carrier distributions of electron and hole are almost symmetry in our simulation. When N_t become larger, because of traps, the hole carrier density become larger and electron density become smaller, which result in larger hole current as discussed before. This effect reduce the difference between hole and electron density near the cathode, and the carrier density become more balanced. The carrier density is more imbalanced near anode, because of electron carrier density decreased, and that is why recombination near the anode is lowing. Fig. 14 shows distribution of recombination rate with $N_t = 10^{18}$cm^{-3}. $\mu 0$ is 10^{-6} cm^2/Vs and E_0 is 10^5 V/cm. The thickness L is 100 nm, $n_0 = 10^{21}$ cm^{-3}, T, T_t are 300 and 1500 K. The figure shows that applying more voltage, electrons are pushed out from the cathode to anode, resulting in the distribution being more smooth. This feature explain why the efficiency E_{ff} become larger with larger applied voltage in literatures [19].

In this section, we also consider the time scale of bipolar device. Fig. 15 shows when we apply larger voltage, the time scale of device reaching the steady state is decreased. $\mu 0$ is 10^{-6}cm^2/Vs and E_0 is 5×10^5V/cm. The temperatures T and T_t are 300 and 1500 K, device thickness is 100 nm. Electrodes are Ca and Au. This Fig. also represents the fact that if trap density is large, device always needs more time to arrive in steady state. Fig. 16 represents the Time-Nt curves with parameters are the same as before.

It is interesting that the time scale in Fig. 6 is smaller than time scale of Fig. 16. We want to know why they are different. In Fig. 17, we compare the time scale of devices

with Ca/Ca and Ca/Au electrodes. They are electron only and bipolar devices. $\mu 0$ is $10^{-6}\mathrm{cm}^2/\mathrm{Vs}$, $n_0 = 10^{21}$ cm^{-3}, and E_0 is $5 \times 10^5 \mathrm{V/cm}$. The thickness is 100 nm and T, T_t are 300 and 1500 K. Total trap density N_t is $10^{17}\mathrm{cm}^{-3}$. We observe the time scale of Ca/Ca device is always faster than that in Ca/Au device in voltages we considered. When voltage is small, the difference between them is large, but when voltage is 8 volt, the difference becomes almost zero. We interpret these phenomena by two reasons. The first, built-in voltage of Ca/Ca is zero. That is, if we apply 2 volt bias, the bias is completely applied on the device. In contrast, if we apply 2 volt bias on Ca/Au device, zero voltage will be applied on device in our consideration(because Ca/Au electrodes have -2 volt built-in voltage). As a result of built-in voltage, the electron current in device with Ca/Ca electrodes is larger than that in Ca/Au device, which result in the difference in time scale. However, bipolar device has hole current, and it will contribute to the electron current injection because of Coulomb effect. In larger voltage, the hole concentration is large enough, so as to compensate for the difference of built-in voltage. That is the reason why the difference between them is reducing with voltage raised.

Fig. 18 shows the Time-Nt curves of devices with Ca/Au and Ca/Al electrodes. In our consideration, the barrier for electron injection to MEH-PPV from Al is 1.4 eV and for hole injection is 0.6 eV. $n_0 = 10^{21}$ cm^{-3}, $\mu 0 = 10^{-6}\mathrm{cm}^2/\mathrm{Vs}$ and $E_0 = 5 \times 10^5 \mathrm{V/cm}$. The thickness $L = 100$ nm and T, T_t are 300, 1500 K. N_t is $10^{17}\mathrm{cm}^{-3}$. This figure shows that the time scale of device with Ca/Au electrodes is faster than that of device with Ca/Al electrodes if the voltage is larger than 5 volt. It is reasonable because the difference of built-in voltage between them is smaller than 2 volt, so the faster time scale of device with Ca/Au electrodes is possible before 8 volt.

4 Conclusion

We have represented results of a device model with traps for single layer organic PLEDs which includes charge injection, transport, and recombination. We investigate the role of traps in devices with space-charge limited current density in which the current is bulk limited. In electron only devices, we investigate the role of traps in electron transport. We also compare our trap model with PF model to find out the differences between them. In bipolar devices, we investigate the trap effect in carrier transport and recombination. Besides, we discuss the effect of trap in time scale of electron only and bipolar devices.

In electron only devices, traps strongly affect the transport of electrons, and the phenomenon that more field dependence in electron current is represented in Fig. 5. Although the PF model can fit the current calculated in trap model, the electron distribution are different between them. Electron density calculated in trap model is less than that calculated in PF model . Quenching effect is important if the major recombination zone being near the cathode, and the PF model is insufficient to discuss the electron distribution through device. Finally, we observed that trap not only suppresses the electron current, but also increase the time scale in which device reach the steady state.

In bipolar devices, we investigate the carrier transport and recombination in devices with traps. We observed that the total current is increasing if the trap density increase. Traps in devices serve as negative space charge centers which contribute to the hole injection and total current. In the condition we considered, the two-times total current density is possible. Because of imbalance between electron and hole density distribution, the efficiency is lowing with more traps, and more traps in devices resulting in recombination lowing near anode and , on the contrary, increasing near cathode. This is a bad news for device performance because of quenching effect. Traps contribute to the increasing of recombination rate near cathode because of balanced electron and hole carrier density. Finally, we observe traps in bipolar devices also affect there time scale. In bipolar devices, hole carrier density can help the electron carrier injection, so they can help the device to reach the steady state faster.

5 Figures

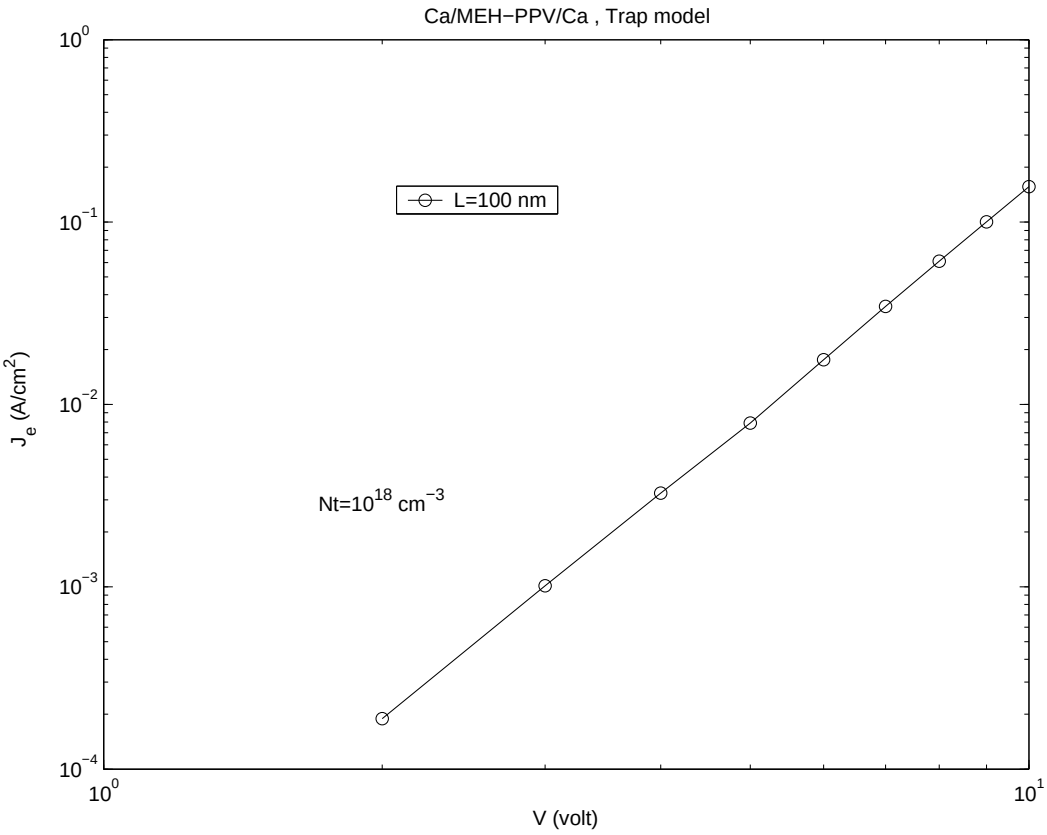


Figure 1: Calculated $J_e - V$ curve of the device with voltages form 2 to 10 volt. We use our trap model with $Nt = 10^{18}$ cm^{-3} and $\mu_0 = 10^{-6}$ cm^2/Vs, $E_0 = 10^5$ V/cm. The thickness of device is 100 nm and the electrodes are Ca. There is a 0.2 eV barrier for electron injection from Ca to MEH-PPV. The temperature T=300 K and $T_t = 1500$ K in consideration.

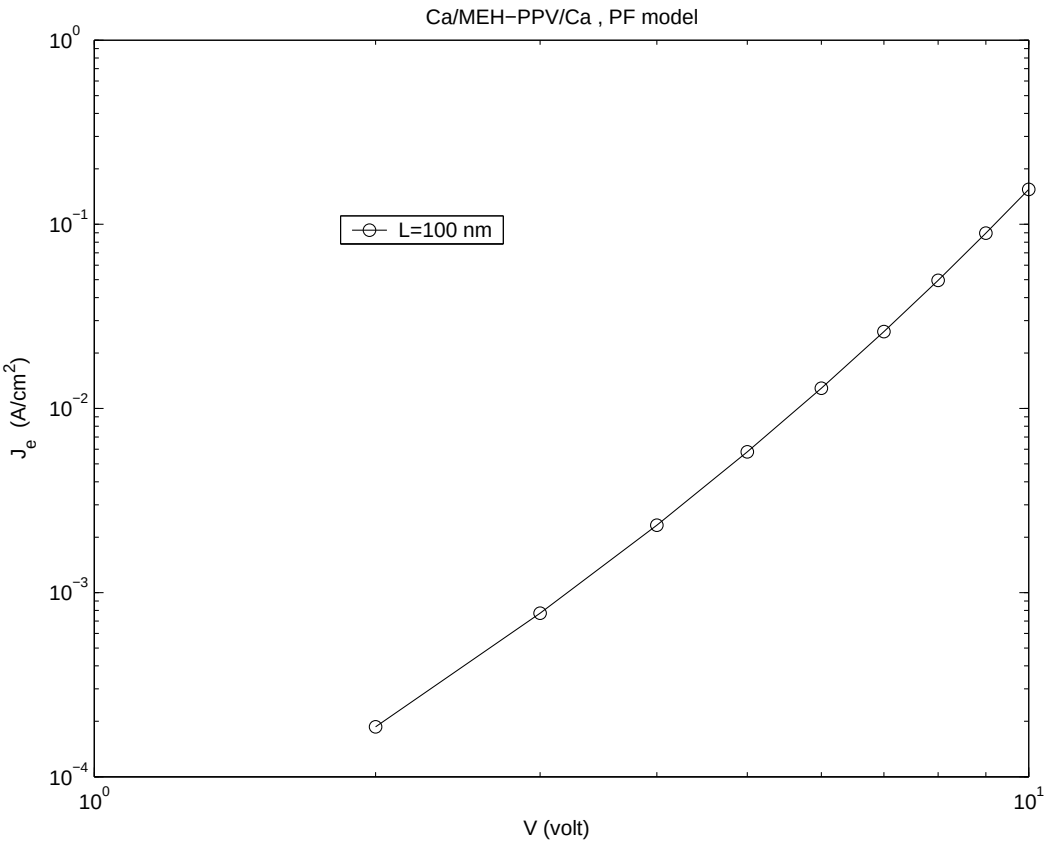


Figure 2: The calculated $J_e - V$ curve of the device using Poole-Frenkel model with $\mu_0 = 8 \times 10^{-}11$ cm^2/Vs and $E_0 = 1.9 \times 10^4$ V/cm in electron mobility. Other parameters are the same as Fig. 1.

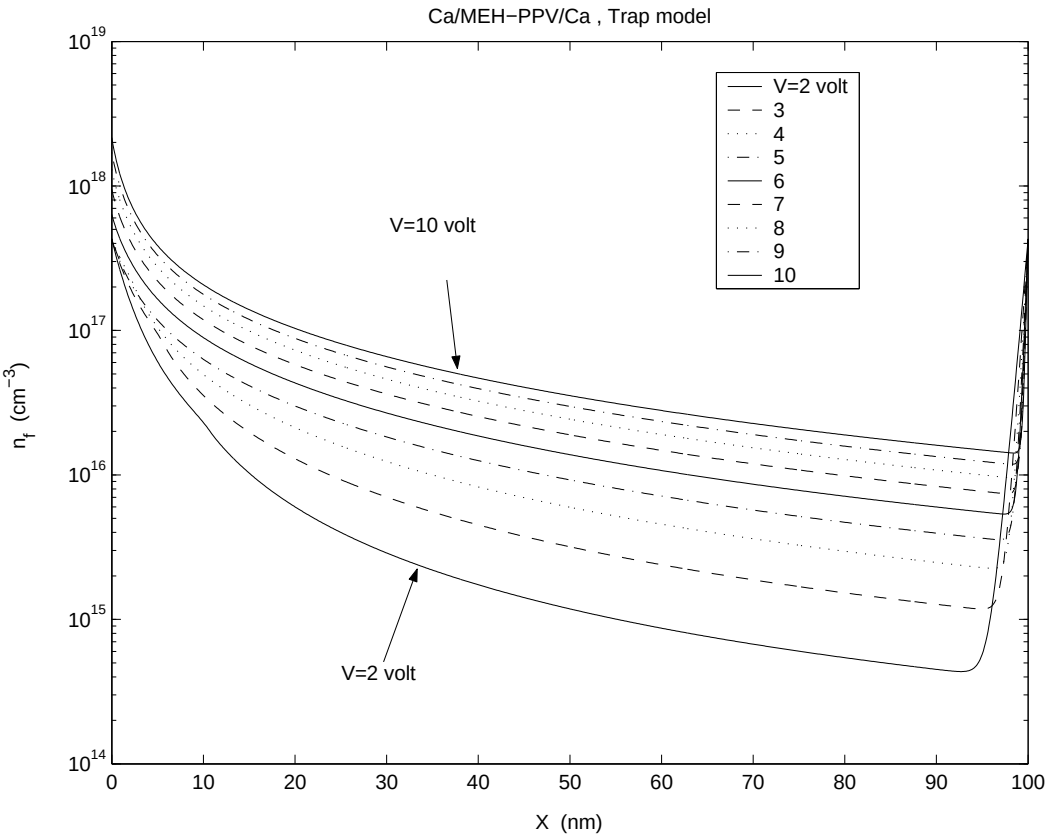


Figure 3: The electron distribution of trap model from applied voltage 2 to 10 volt, and the electrons are injected from left hand side. The thickness is 100 nm, T and T_t are 300 and 1500 K

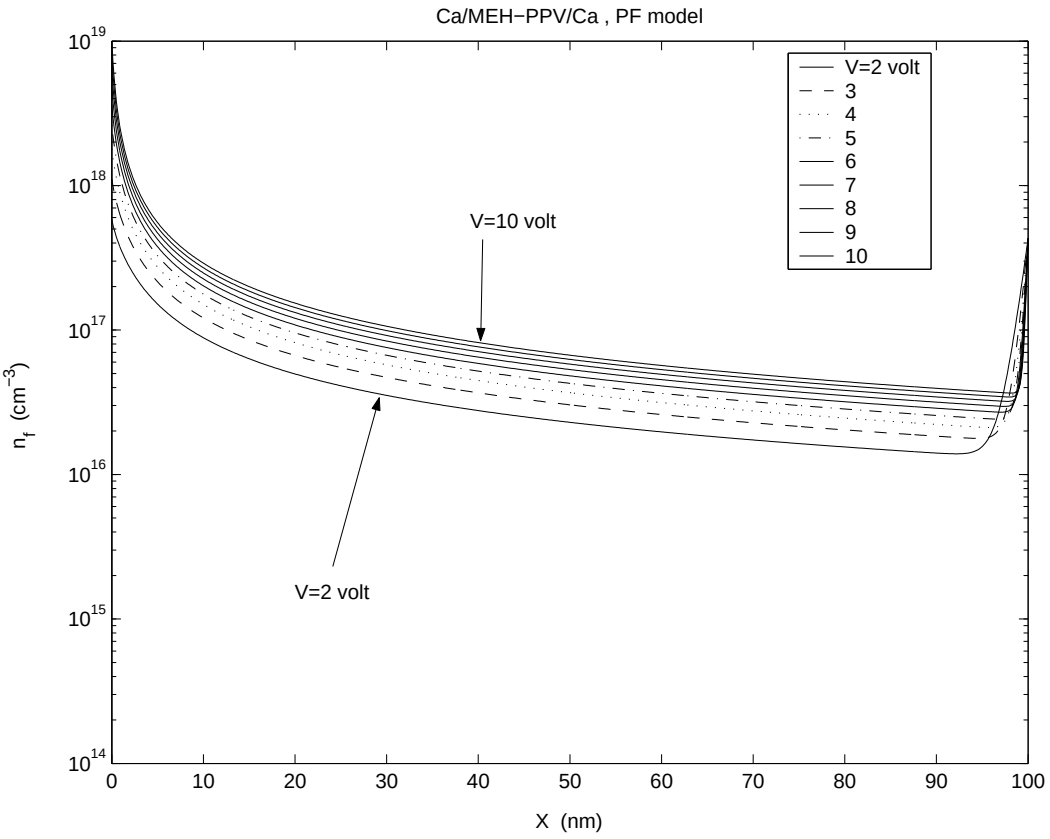


Figure 4: The electron distribution in PF model from applied voltage 2 to 10volt. Using $\mu_0 = 8 \times 10^{-11}$ cm^2/Vs and $E_0 = 1.9 \times 10^4$ V/cm in electron mobility, $L = 100$ nm, $T$and T_t are 300 and 1500 K.

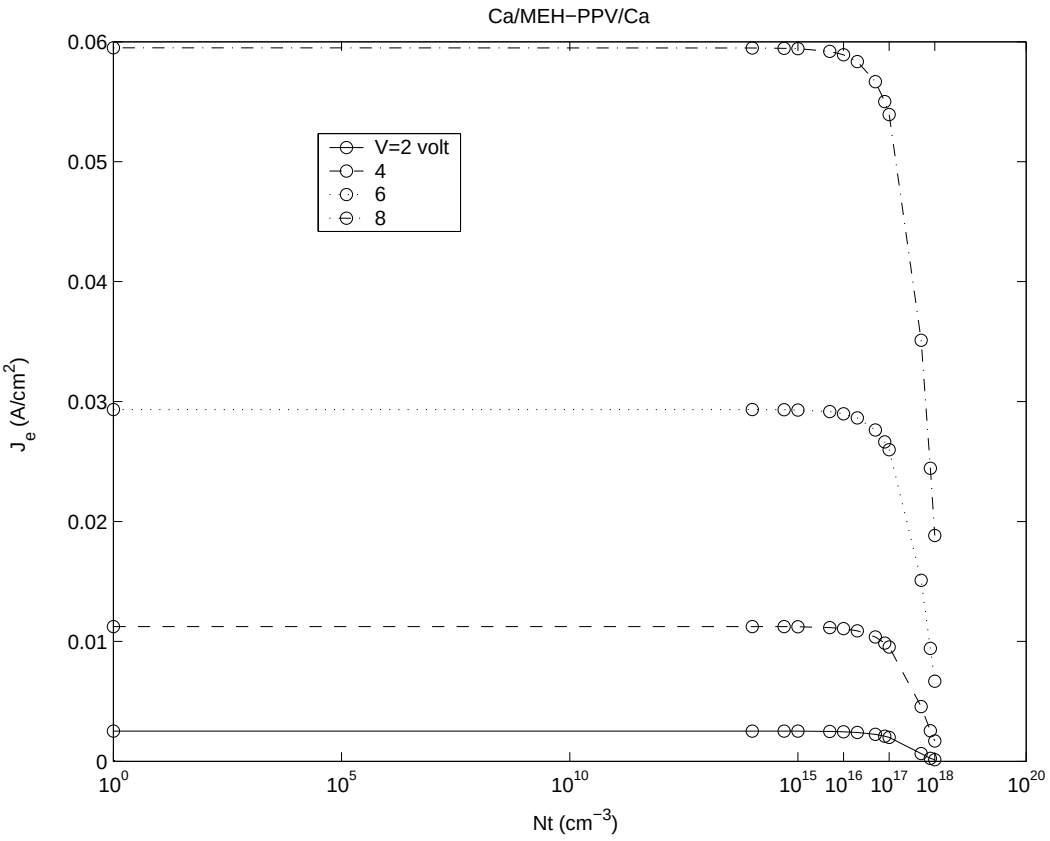


Figure 5: Calculated $J_e - N_t$ curve is shown with voltage 2, 4, 6, and 8 volt. $\mu_0 = 10^{-6}$ cm^2/Vs, $E_0 = 5 \times 10^5$ V/cm, T and T_t are 300 and 1500 K. The thickness L is 100 nm. We observed the electron current is suppressed because of traps.

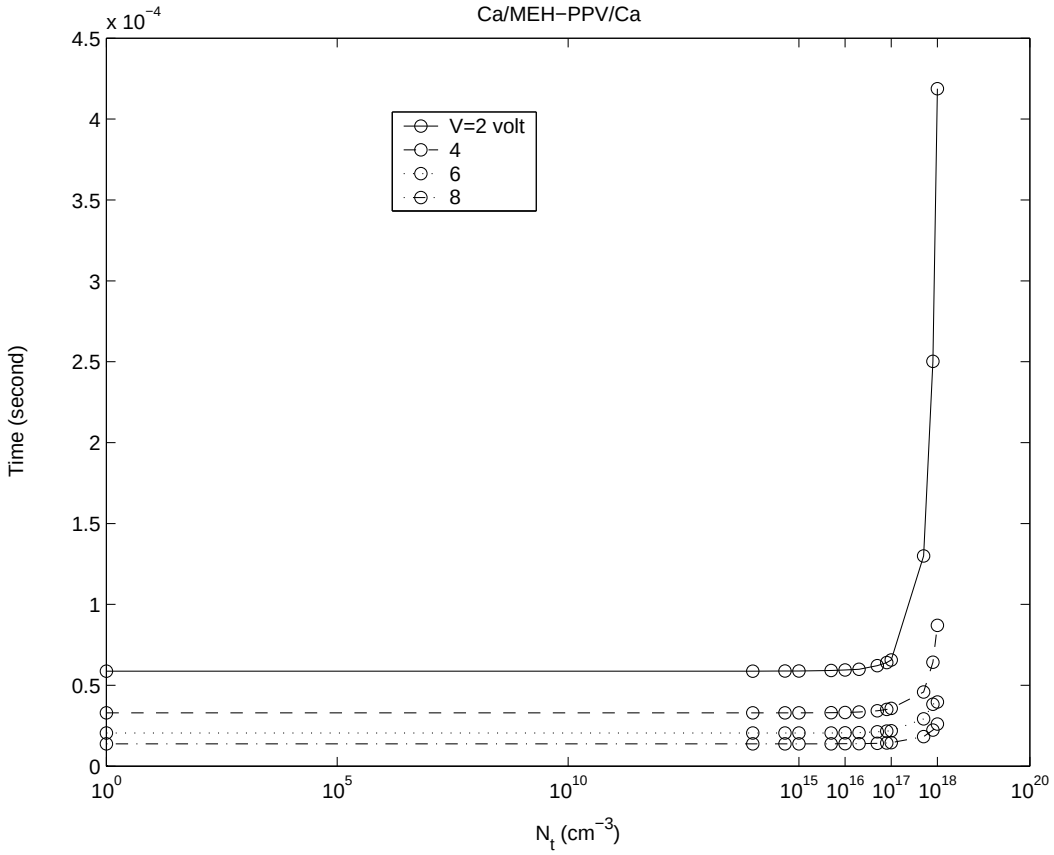


Figure 6: Calculated $Time - N_t$ curve is shown with voltage 2, 4, 6, and 8 volt. $\mu_0 = 10^{-6}$ cm^2/Vs, $E_0 = 5 \times 10^5$ V/cm, T and T_t are 300 and 1500 K. The thickness L is 100 nm. For smaller voltage, device must spend more time arriving at the steady state.

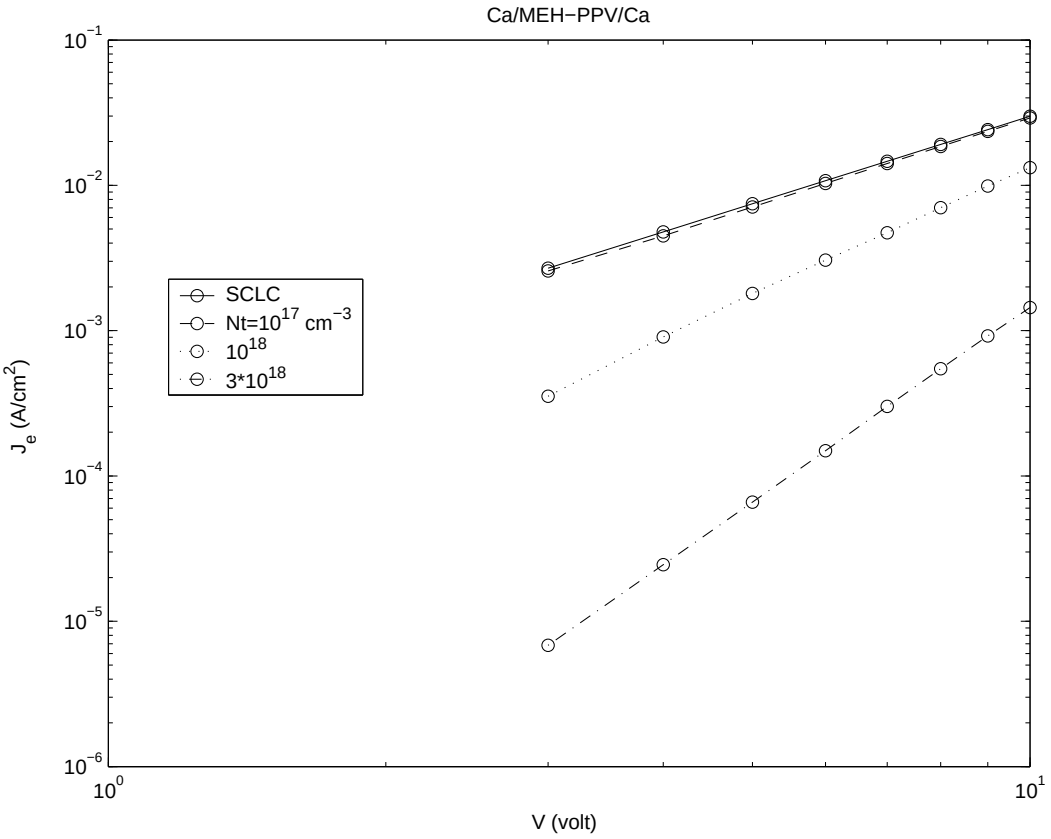


Figure 7: Log-log plot of calculated current density as a function of voltage with trap density 10^{17}, 10^{18}, and $3 \times 10^{18}\mathrm{cm}^{-3}$. μ_0=$10^{-6}\mathrm{cm}^2/\mathrm{Vs}$, n_0=$10^{21}\mathrm{cm}^{-3}$, $E_0 = 10^5$ V/cm. The thickness L is 100nm and T, T_t are 300 and 1500 K. The more field dependence of electron current density is observed. SCLC is considered as trap free current density.

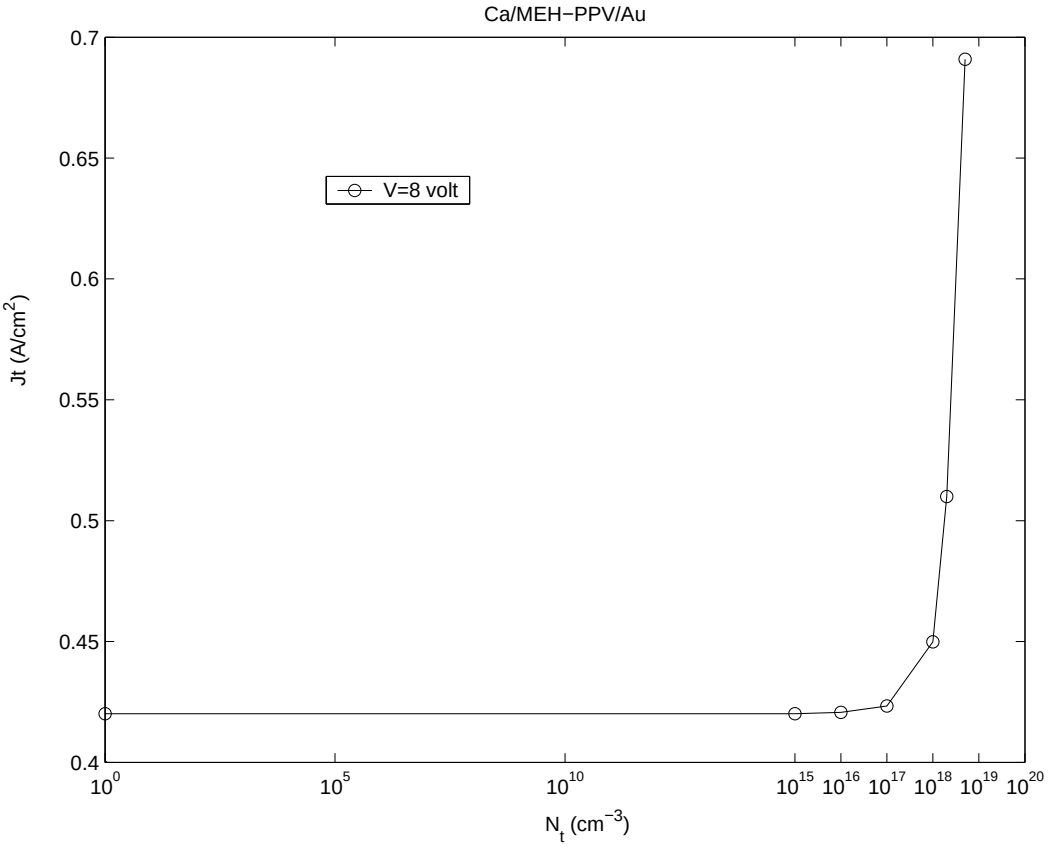


Figure 8: The $J_t - Nt$ curve of Ca/MEH-PPV/Au bipolar device with trap density $1, 10^{15}, 10^{16}, 10^{17}, 10^{18}, 2 \times 10^{18}$, and $5 \times 10^{18}\text{cm}^{-3}$ is shown. Thickness L=100 nm and $V_{bias} = 8$ volt. The parameters n_0, μ_0 and E_0 are 10^{21} cm^{-3}, 10^{-6} cm^2/Vs and 10^5 V/cm. The temperature T and T_t are 300 and 1500 K. It is clear that the more trap density, the more total current density in bipolar devices.

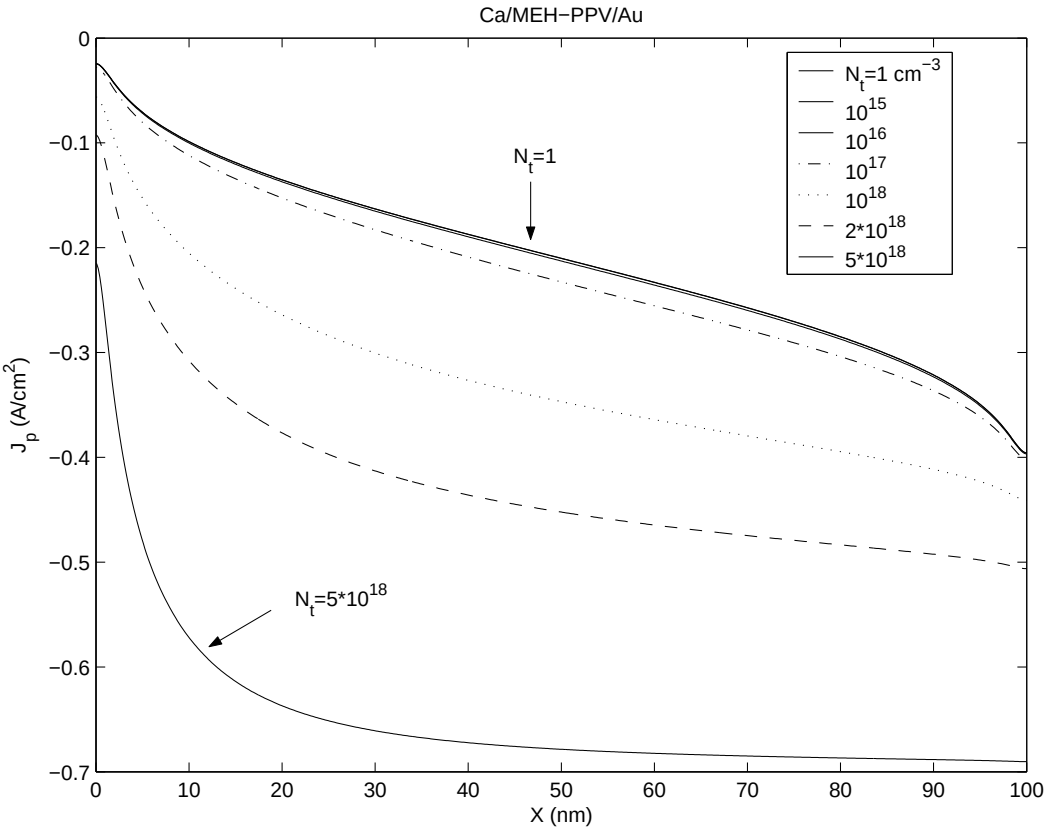


Figure 9: Curves of hole current distribution are represented with parameters are the same as Fig. 8. The injection of hole current increase with increased trap density.

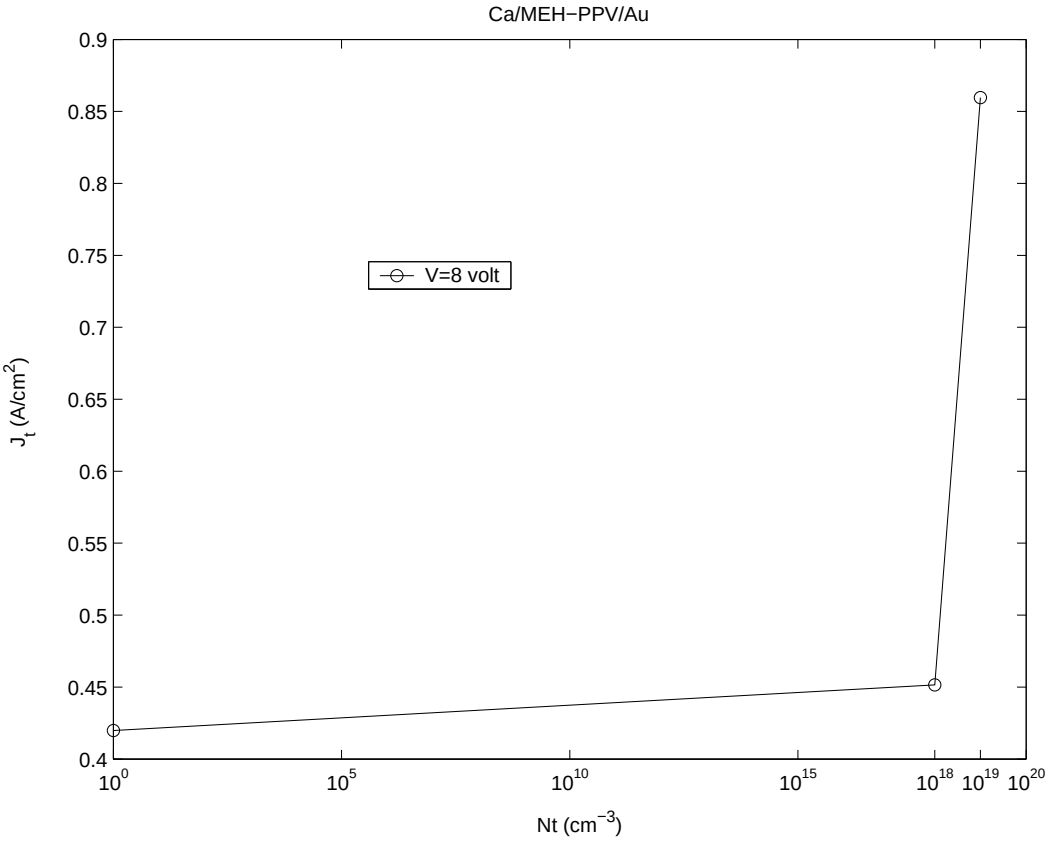


Figure 10: Calculated $J_t - Nt$ curve with V_{bias} = 8volt and thickness L=100 nm is represented. The parameters n_0, μ_0 and E_0 are 10^{21} cm^{-3}, 10^{-6} cm^2/Vs and 10^5 V/cm. The temperature T and T_t are 300 and 1500 K. The total current J_t in trap density $N_t = 10^{19}$ cm^{-3} is almost two-times magnitude larger than that in $N_t = 1$ cm^{-3}.

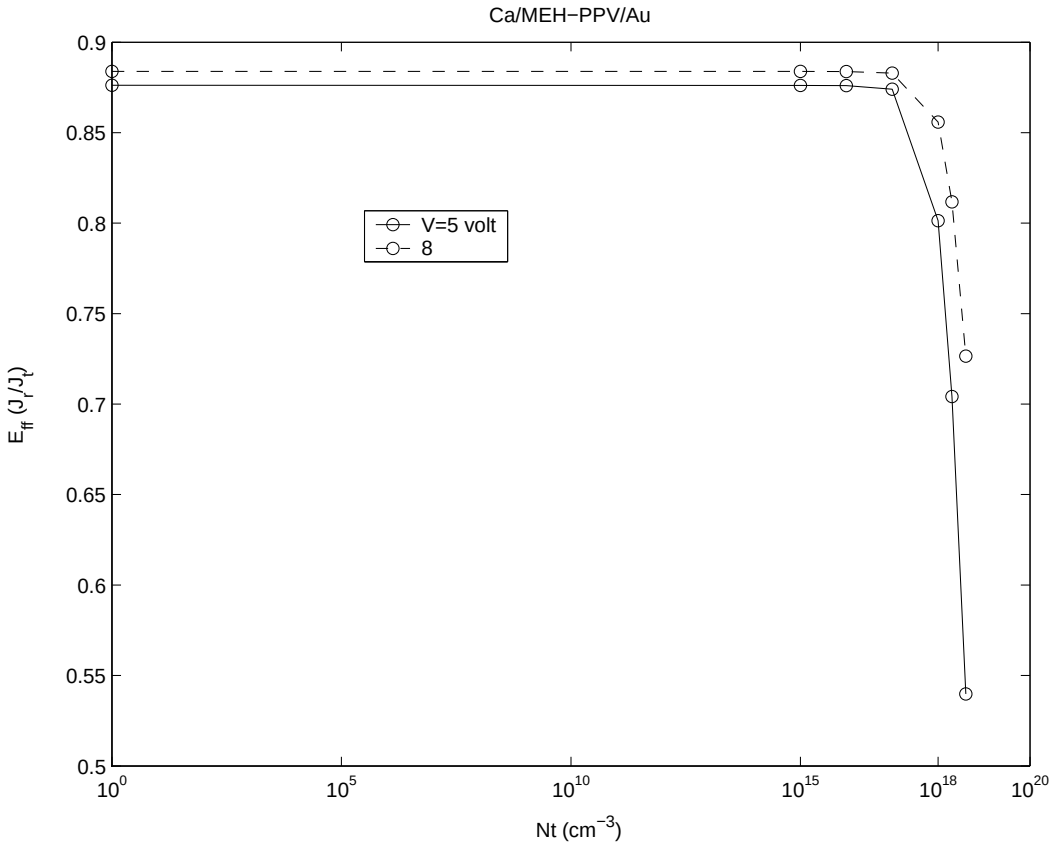


Figure 11: Calculated $E_{ff} - N_t$ curves are represented with bias $V = 5$ and 8 volt. Total trap density N_t varies from 1 to $4 \times 10^{18}\text{cm}^{-3}$. The parameters are the same as Fig. 10. The efficiency with applied bias $V = 5$volt is lowing faster than that with bias 8 volt, showing the fact that the imbalance between electron and hole current in 5 volt is more than that in 8 volt.

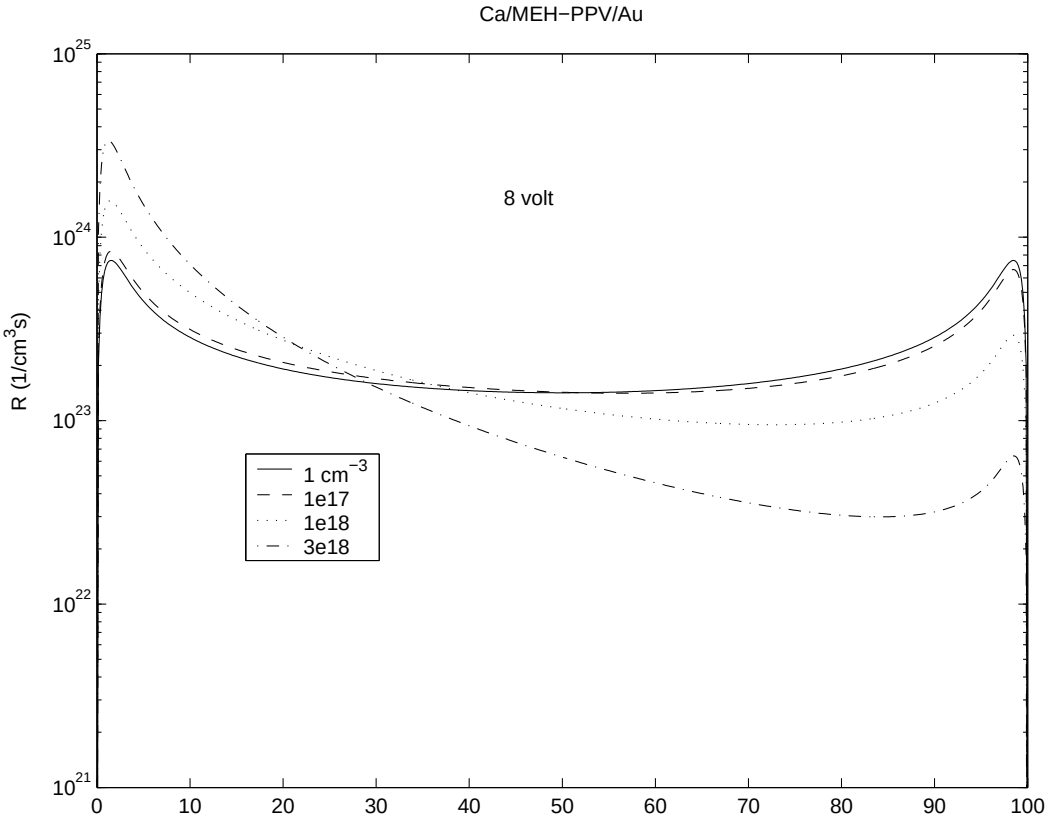


Figure 12: Calculated distribution of the light generation in a PLED. The distributions with trap density below 10^{17} cm^{-3} are almost the same in our simulation. Thickness L=100 nm. The parameters n_0, μ_0 and E_0 are 10^{21} cm^{-3}, 10^{-6} cm^2/Vs and 10^5 V/cm. The temperature T and T_t are 300 and 1500 K. The major recombination zone is approaching to the cathode with increased trap density.

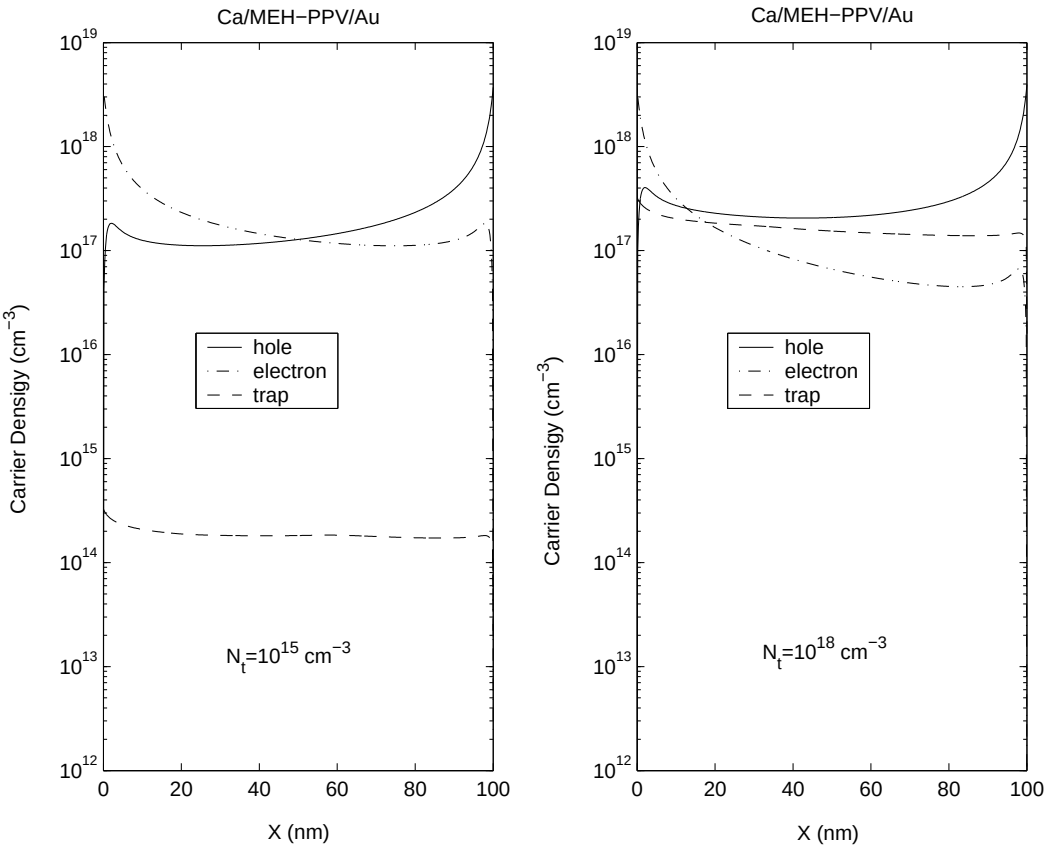


Figure 13: Calculated carrier distribution with $N_t = 10^{15}\text{cm}^{-3}$ and$N_t = 10^{18}\text{cm}^{-3}$ are represented. Because of more trap density, the electron density in $N_t = 10^{18}\text{cm}^{-3}$ is smaller and hole is larger than that in $N_t = 10^{15}\text{cm}^{-3}$, which result in the increasing in recombination near cathode and reduction near anode. Left hand side is cathode and right hand side is anode.

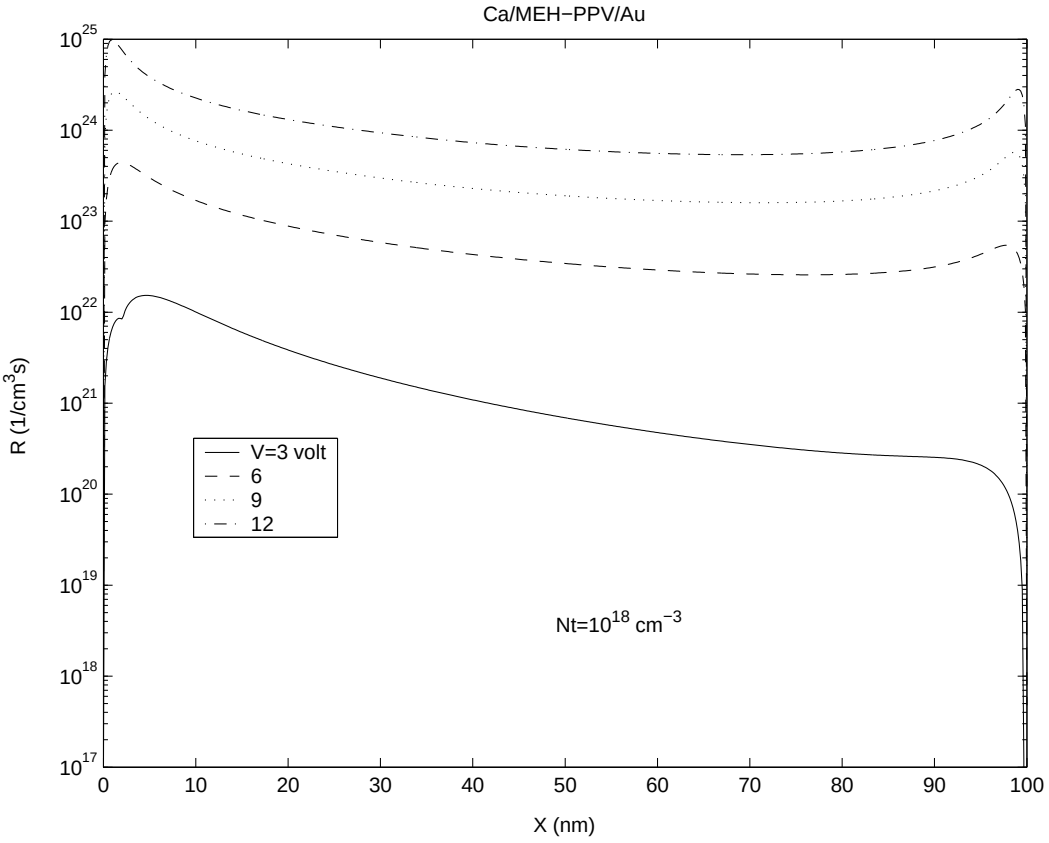


Figure 14: Distribution of recombination rate are calculated. The bias varies from 3 to 12 volt in our consideration. We consider the recombination rate with trap density $10^{18}\mathrm{cm}^{-3}$. Other parameters are the same as in Fig. 12. This figure represents the phenomenon that when we apply larger bias, more smooth the distribution is.

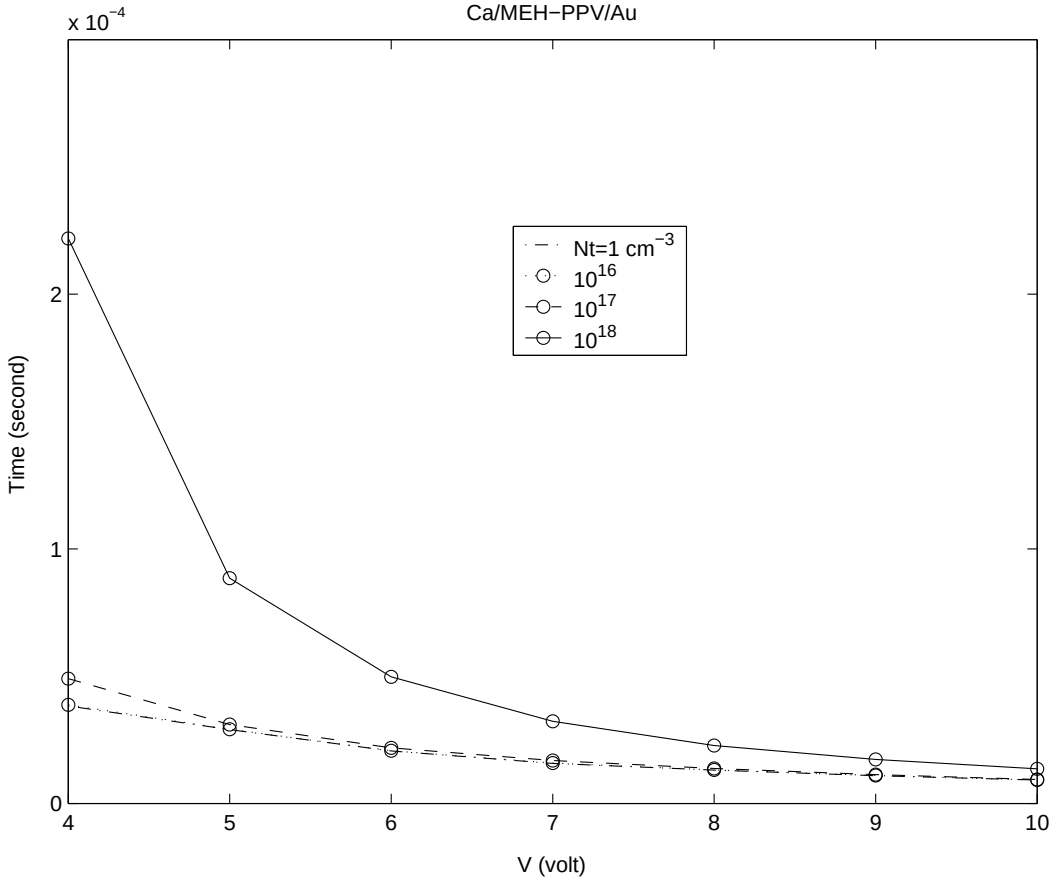


Figure 15: Time scale of device is represented. $\mu 0$ is $10^{-6}\mathrm{cm}^2/\mathrm{Vs}$ and E_0 is $5 \times 10^5 \mathrm{V/cm}$. The temperatures T and T_t are 300 and 1500 K, device thickness is 100 nm, n_0 is $10^{21}\mathrm{cm}^{-3}$. The curves of trap density 1 and 10^{16} cm^{-3} are almost the same in our simulation. Larger voltage needs less time to fill the trap, so the time scale is small.

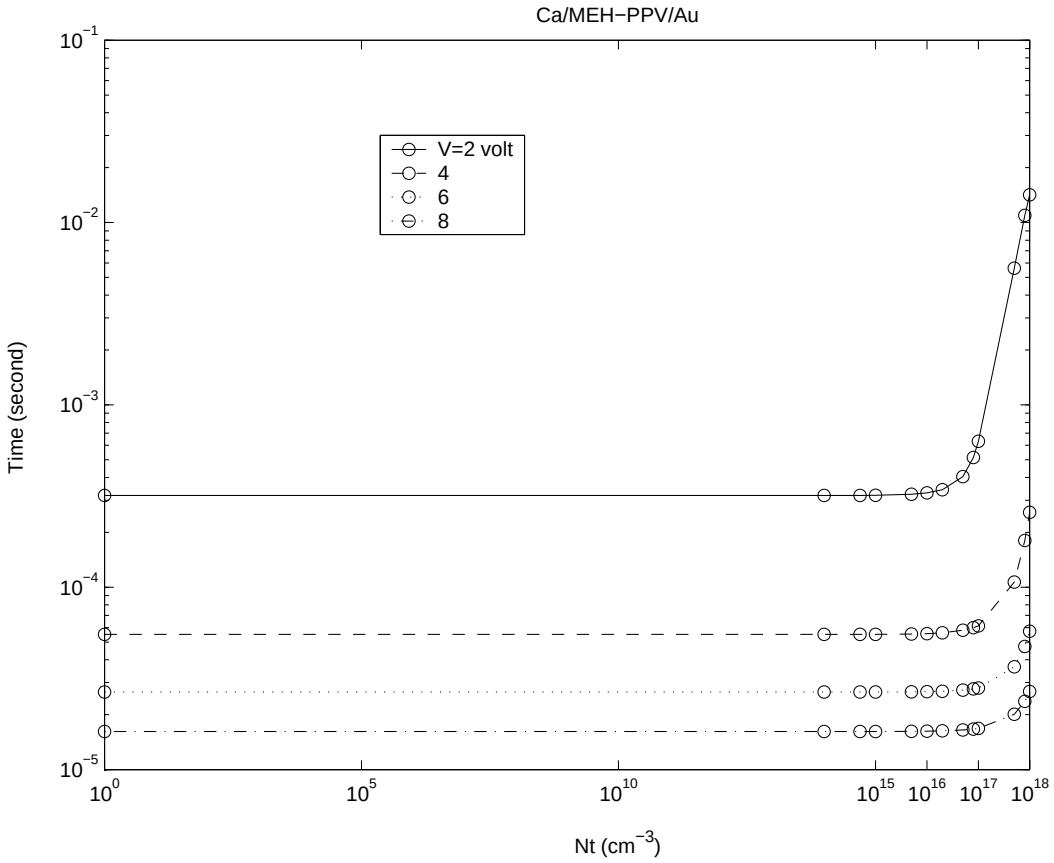


Figure 16: The Time-Nt curves are represented. The curves with voltages are 2, 4, 6 and 8 volt. Other parameters are the same as before figure. Device with smaller voltage needs more time to reach steady state, and if the trap density become large enough, trap will affect the time scale greatly.

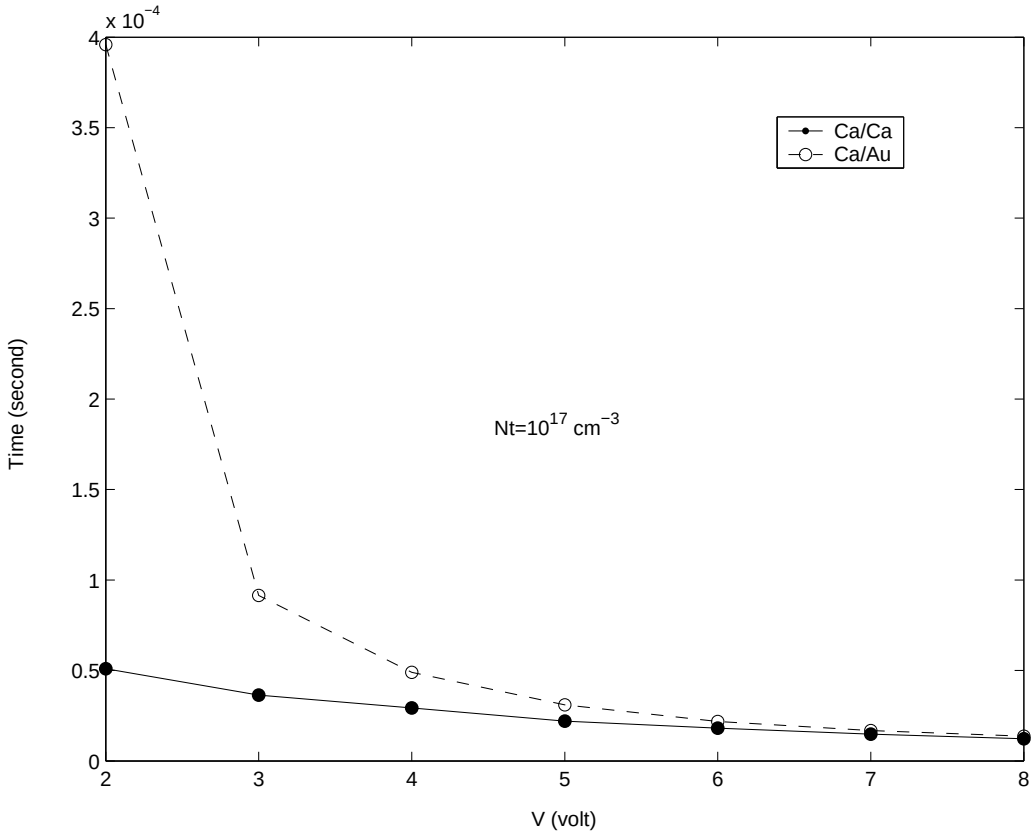


Figure 17: The Time-Nt curves of Ca/Ca and Ca/Au electrodes are considered, and voltages varied from 2 to 8 volt. The trap density we considered is 10^{17} cm^{-3}. $\mu 0$ is 10^{-6}cm^2/Vs and E_0 is 5×10^5V/cm. The temperatures T and T_t are 300 and 1500 K, device thickness is 100 nm, n_0 is 10^{21}cm^{-3}. Because of built-in voltage, the time scale of Ca/Au device with voltages below 8 volt is bigger than that in Ca/Ca voltage.

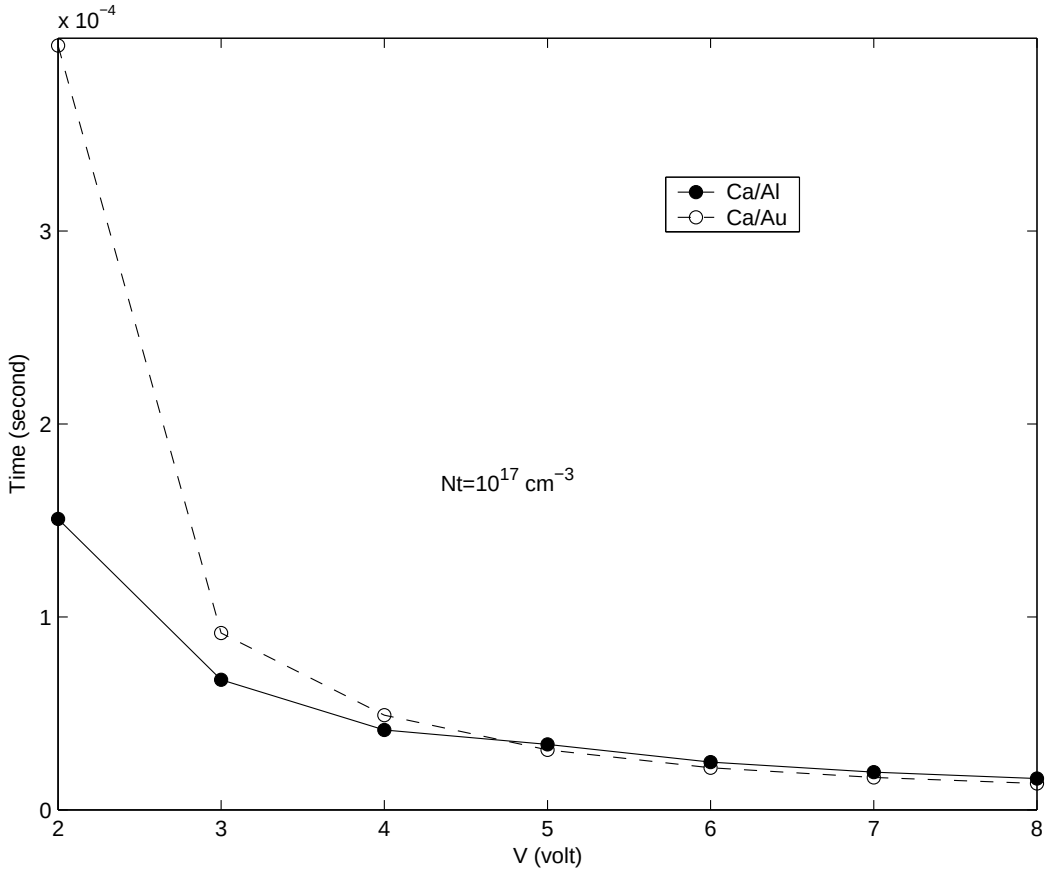


Figure 18: Time-Nt curves of Ca/Al and Ca/Au electrodes are represented. We use $\mu_0 = 10^{-6}\text{cm}^2/\text{Vs}$ and E_0=5×10^5 V/cm. T, T_t are 300 and 1500 K, thickness L=100 nm. Total trap density Nt is 10^{17}cm^{-3}. We observe that the time scale of device with Ca/Au electrodes is faster than device with Ca/Al electrodes when voltage is larger than 5 volt.

6 Appendix

A Time Derivative of Electric Field

We take the time derivative of Poisson's equation:

$$\frac{\partial}{\partial t}(\frac{\partial E}{\partial x} = \frac{4\pi e}{\epsilon}(p-n)) \tag{34}$$

$$\frac{\partial\partial}{\partial x \partial t}E = \frac{4\pi e}{\epsilon}\frac{\partial}{\partial t}(p-n) = -\frac{4\pi}{\epsilon}\frac{\partial}{\partial x}J_t. \tag{35}$$

Hence,

$$\frac{\partial}{\partial x}(\frac{\partial}{\partial t}E + \frac{4\pi}{\epsilon}J_t) = 0 \tag{36}$$

$$\Rightarrow \frac{\partial}{\partial t}E + \frac{4\pi}{\epsilon}J_t = constant\ with\ respect\ to\ x. \tag{37}$$

In order to get the value of constant, we integrate the equation (31) and take the average value, that is

$$\frac{\partial}{\partial t}E + \frac{4\pi}{\epsilon}J_t = \frac{1}{L}\int_0^L (\frac{\partial}{\partial t}E + \frac{4\pi}{\epsilon}J_t)dx. \tag{38}$$

Here we introduce the boundary condition $\phi(0) = 0$ and $\phi(L) = V_L$, hence

$$\frac{\partial}{\partial t}E = -\frac{1\partial}{L\partial t}V_L - \frac{4\pi}{\epsilon}[J_t(x) - \frac{1}{L}\int_0^L J_t(x)\ dx]. \tag{39}$$

Consequently, we use this equation to find out the steady state solution of electric field.

B Time Derivative of Free and Trapped Electron Carrier Density

Here, we want to find out the time derivative of n_f and n_t. First of all, we need the trap density:

$$n_{t\varepsilon}(\varepsilon) = \frac{N_t}{kT_t} e^{(\varepsilon-(\varepsilon_c-eV))\beta} \tag{40}$$

where $\beta_1 = 1/kT_t$. So we have

$$n_t(x) = \int_{-\infty}^{\infty} n_{t\varepsilon}(\varepsilon) f d\varepsilon \tag{41}$$

where f is Fermi-Dirac distribution. Here, because the kT_t (0.15eV)is larger than kT(0.0259 eV) in room temperature, that is, we can assume the Fermi-Dirac distribution is a step function. So, we have

$$n_t(x) \simeq \int_{-\infty}^{\mu} \frac{N_t}{kT_t} e^{(\varepsilon-(\varepsilon_c-eV))\beta_1} d\varepsilon \tag{42}$$

$$= \frac{N_t}{kT_t} e^{-(\varepsilon_c-eV)\beta} \int_{-\infty}^{\mu} e^{\varepsilon\beta_1} \tag{43}$$

where μ is the local chemical potential. Now, we need the chemical potential to find the trap density. Because the trapped electrons are electrons scattering with phonons, and the time scale ($10^{-12}sec$) is smaller than that of device reaching steady state($10^{-6}sec$), we assume the free electron and trapped electron are locally in thermal equilibrium. So we use the thermal equilibrium carrier density

$$n_f = n_0 e^{-(\varepsilon-\mu-eV)\beta} \tag{44}$$

where $\beta = 1/kT$. Therefore

$$\ln \frac{n_f}{n_0} = -\beta(\varepsilon - \mu - eV) \tag{45}$$

$$\Rightarrow \mu = \varepsilon - eV + \frac{1}{\beta} \ln \frac{n_f}{n_0}. \tag{46}$$

Take the μ into equation (44)

$$\Rightarrow n_t(x) = N_t e^{-(\varepsilon_c-eV)\beta_1} e^{\mu\beta_1} \tag{47}$$

$$= N_t e^{\frac{\beta_1}{\beta} \ln \frac{n_f}{n_0}} \tag{48}$$

$$= N_t (\frac{n_f}{n_0})^{T/T_t}. \tag{49}$$

Now, we take the time derivative of total electron density

$$\frac{\partial}{\partial t} n_{to} = \frac{\partial}{\partial t} n_f + \frac{\partial}{\partial t} n_t \tag{50}$$

$$= \frac{\partial}{\partial t} n_f + N_t (\frac{1}{n_0})^{T/T_t} \frac{T}{T_t} n_f^{T/T_t - 1} \frac{\partial}{\partial t} n_f \tag{51}$$

$$\equiv (1 + B) \frac{\partial}{\partial t} n_f \tag{52}$$

where

$$B = N_t \frac{T}{T_t} (\frac{1}{n_0})^{T/T_t} (\frac{1}{n_f})^{1 - T/T_t}.$$

Therefore,

$$(1 + B) \frac{\partial}{\partial t} n_f = \frac{1}{e} \frac{\partial}{\partial x} J_n - r n_f p \tag{53}$$

$$\Rightarrow \frac{\partial}{\partial t} n_f = \frac{1}{1 + B} [\frac{\partial}{\partial x} J_n - r n_f p] \tag{54}$$

and

$$\frac{\partial}{\partial t} n_t = \frac{B}{1 + B} [\frac{\partial}{\partial x} J_n - r n_f p]. \tag{55}$$

C Space Charge Limited Current(SCLC)

SCLC describe the current of organic device in bulk limited regime. Because of the low carrier mobility in organic material, the carriers in device will affect the electric field through the device that result in the bulk limited current density. We consider the hole current density and neglect the diffusion current. In steady state,

$$\frac{1}{e}\frac{\partial}{\partial x}J_p = 0.$$

J_p is constant with respect to x. J_p is given by

$$J_p = ep\mu E,\ p = J_p/e\mu E.$$

Therefore, the Poisson's equation can be written as

$$\frac{\partial}{\partial x}E = \frac{e}{\epsilon_0\epsilon_r}p = \frac{1}{\mu\epsilon_0\epsilon_r}\frac{J_p}{E}.$$

The phenomenon of SLCL arises when the contacts inject too much charge to transport. In this case the current is determined by transport property of device. We have

$$EdE = \frac{1}{\mu\epsilon_0\epsilon_r}J_p\ dx$$

$$\Rightarrow \frac{1}{2}E^2 = \frac{1}{\mu\epsilon_0\epsilon_r}J_p\ x$$

$$\Rightarrow V = \int_0^L Edx = \frac{2}{3}(\frac{2}{\mu\epsilon_0\epsilon_r}J_p)^{1/2}L^{3/2}.$$

So we have

$$J_p = \frac{9}{8}\mu\epsilon_0\epsilon_r\frac{V^2}{L^3}.$$

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