

Diffusion-controlled charge injection model for organic light-emitting diodes

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We present an compact model to describe the charge injection for organic light-emitting diodes. By identifying a critical distance where the concentration of carriers in the extended states equals that of the trapped carriers, we obtain a model for the injection current, which links the drift-diffusion and the multiple-trapping theories. This model yields the injection current as a function of electric field, temperature, and barrier height between metal and organic semiconductor. Good agreement with recent experimental data is observed. The effect of the field-dependent mobility on the injection current is also discussed. © 2007 American Institute of Physics.

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Over the past 10 years, there has been a surge of interest in the development and application of organic semiconductors, such as organic light-emitting diodes (OLEDs) and organic field effect transistors.^{1,2} However, on the theoretical side, there is still a lack of satisfactory description of the physical process underlying the charge injection in organic light-emitting diodes. For example, the Fowler-Nordheim model for tunneling injection and the Richardson-Schottky model for thermionic emission³ were developed for regular band-type semiconductor materials. They are, however, insufficient to handle disordered organic materials, where the density of states (DOS) is a Gaussian distribution, with localized charge carriers and discrete hopping within a distribution of energy states.⁴ Arkhipov *et al.* presented an analytical model based on hopping theory,⁵ and Wolf *et al.* performed detailed Monte Carlo simulations of charge injection from a metal to an organic semiconductor layer.⁶

Due to the low mobility in organic semiconductors ($\mu \ll 10^{-3} \text{ cm}^2/\text{V s}$), the diffusion transport is also very important for the charge injection process. At the same time, different parameters such as barrier height, mobility, and device length affect the current in OLED. It is useful to consider an organic diode structure in which a single carrier-type (i.e., either electrons or holes) dominates the current flow in order to clarify the device operation in a relatively simple situation.⁷ Such devices can be easily fabricated by choosing the contact so that the energy barrier for one carrier injection is much higher than that for the other. Furthermore, since that the electron mobility (μ_e) is much larger than the hole mobility (μ_h) in organic semiconductors [$\mu_h \approx 0.01\mu_e$ (Ref. 8)], it is commonly believed that the electrons would travel all the way to the interface between the electron transport layer and hole transport layer and recombine with holes in the immediate vicinity of that interface. So the double carrier injection current should be approximately equal to the single carrier injection current for some OLEDs with thicker organic film and higher hole injection barrier.^{9,10} Therefore, the aim of this work is to develop an analytical, diffusion-controlled single carrier charge injection model particularly suited for OLED. This model is based on drift-diffusion and multiple trapping theories. The latter can be used to describe

hopping transport in organic semiconductors.¹¹ The presented model can be used to explain the dependence of the injection current on the temperature, the electric field, and the barrier height. The theoretical predictions agree well with the experimental data.

The present work is concerned with injection-limited conduction at high electric field. The potential barrier $e\varphi(x)$, formed at the metal semiconductor interface, is a superposition of an external electric field and a Coulomb field binding the carrier on the electrode,^{12,13}

$$e\varphi(x) = \Delta - \frac{e^2}{16\pi\epsilon_0\epsilon x} - eFx. \quad (1)$$

Here, x is the distance to interest and the metal-organic layer interface. Since the rapid variation of the potential Eq. (1) takes place in front of the cathode, the field F can be regarded as being nearly constant.

Using the drift-diffusion theory, the hole current J can be written as

$$-J = k_B T \mu \left[\frac{e}{k_B T} p_e(x) \frac{d\varphi(x)}{dx} + \frac{dp_e(x)}{dx} \right], \quad (2)$$

where k_B is the Boltzmann constant, $p_e(x)$ is the hole concentration in the extended states, and μ is the mobility. On taking J and μ as constant, and solving for $p_e(x)$, we obtain

$$p_e(x) = \left[N - \frac{J}{k_B T \mu} \int_0^x \exp\left(\frac{e\varphi(x')}{k_B T}\right) dx' \right] \exp\left(-\frac{e\varphi(x)}{k_B T}\right), \quad (3)$$

where N is the carrier concentration at $x=0$. In multiple trapping theory,¹⁴ the total carrier concentration is given by a sum of carrier concentrations in the extended states $p_e(x)$ and the localized states,

$$p(x) = p_e(x) + \int_0^\infty g(E, x) f(E, E_F) dE. \quad (4)$$

Here, $g(E)$ is the density of the localized states, $f(E, E_F)$ is the Fermi Dirac distribution, and the quasi-Fermi energy E_F can be written as

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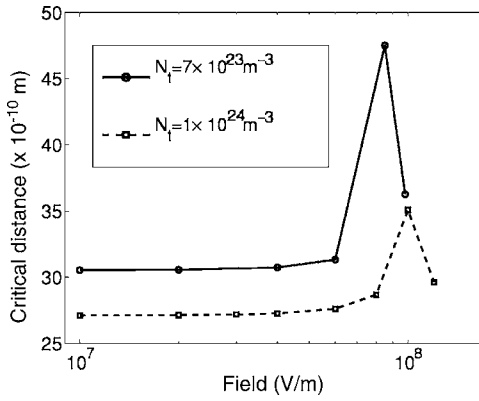


FIG. 1. Dependence of the critical distance on the electric field.

$$E_F(x) = k_B T \ln \left[\frac{\nu_0 \tau_0 N_t}{p_e(x)} \right],$$

where N_t is the total concentration of localized states, τ_0 is the lifetime of carriers, and ν_0 is the attempt-to-escape frequency.

In the injection regime, very close to the contact all the traps are filled. Moreover, the carrier concentration in the extended states is much higher than that in the trapped states. At large distance from the injection contact, the main contribution to the total carrier concentration comes from the occupied localized states.⁵ So we propose here the concept of *critical distance* x_d , where the carrier concentration in the extended states equals the carrier concentration in the localized states, i.e.,

$$p_e(x_d) = \int_0^\infty g(E, x_d) f(E, E_F) dE. \quad (5)$$

Substituting Eqs. (1), (4), and (5) into the Poisson equation,

$$\frac{d^2(e\phi)}{dx^2} = -\frac{e}{\epsilon_0 \epsilon} p(x), \quad (6)$$

the critical distance x_d can be calculated from

$$1 = \int_0^\infty \frac{16\pi x_d^3 g[E - e\phi(x_d)]}{1 + 16\pi x_d^3 \nu_0 \tau_0 N_t \exp(-E/k_B T)}. \quad (7)$$

In various disordered organic systems, a pure Gaussian DOS can be assumed,

$$g(E) = \frac{N_t}{\sqrt{2\pi}\sigma} \exp\left(-\frac{E^2}{2\sigma^2}\right), \quad (8)$$

where σ is the standard deviation. Solving Eqs. (7) and (8) numerically, we can obtain the critical distance x_d . The free carrier concentration at x_d is calculated by Eq. (5). Finally, the injection current can be calculated as

$$J = k_B T \mu \frac{\left[N - p_e(x_d) \exp\left(\frac{e\phi(x_d)}{k_B T}\right) \right]}{\int_0^{x_d} \exp\left(\frac{\phi(x)}{k_B T}\right) dx}. \quad (9)$$

The critical distance as a function of the electric field is plotted in Fig. 1 for different N_t . The parameters are $\sigma/k_B T = 4$, $\Delta = 0.4$ eV, $\nu_0 = 1 \times 10^{12}$ s⁻¹, and $\tau_0 = 1 \times 10^{-12}$ s. We can see that the critical distance decreases with N_t . However, the

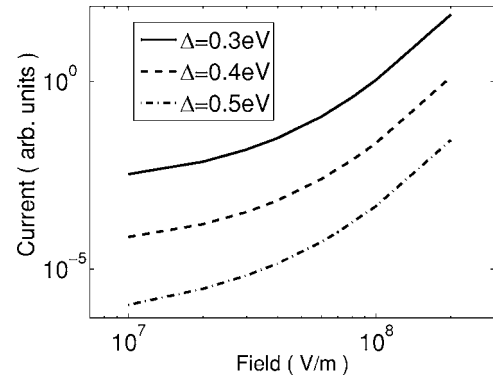


FIG. 2. Dependence of the injection current on the barrier height.

effect of the electric field is non monotonic. The critical distance increases with electric field in the lower electric field regime, and decreases with electric field in the higher electric field.

The barrier height Δ plays an important role in injection efficiency. We calculate the relation between the injection current and the electric field for different Δ , as shown in Fig. 2. The parameters are $N_t = 1 \times 10^{18}$ cm⁻³, $\sigma = 0.1656$ eV, $\nu_0 = 1 \times 10^{11}$ s⁻¹, $\tau_0 = 1 \times 10^{-11}$ s, $T = 300$ K, and $\mu = 1 \times 10^{-9}$ cm²/V s. The injection current increases with the electric field, and the lower the Δ , the higher the injection current as intuitively expected. However, the slope of $\log J$ vs \log is not constant.

Figure 3 shows the temperature dependence of the injection current with $\Delta = 0.3$ eV, where other parameters are the same as in Fig. 2. The temperature coefficient decreases strongly with increasing electric field. The coefficient reverses sign at high electric field, which has also been observed in Ref. 13 theoretically.

A comparison between the model prediction and the experimental data⁵ is shown in Fig. 4 $\Delta = 0.3$ eV and $\Delta = 0.5$ eV. The fitting parameters are $N_t = 1 \times 10^{17}$ cm⁻³, $\mu = 2.56 \times 10^{-11}$ cm²/V s for PPV-ether, and 2.51×10^{-9} cm²/V s for PPV-imine, respectively. The other parameters are the same as in Fig. 2.

Next, we compute the charge injection for the OLED using our model assuming a field-dependent mobility. Note that there have been well-known models for charge injection in OLEDs with constant mobility. However, the mobility in organic materials depends on the local electric field F as¹⁵

$$\mu(F) = \mu_0 \exp(\gamma\sqrt{F}). \quad (10)$$

Here, μ_0 denotes the mobility of carriers at zero field and γ is the parameter describing the field dependence. We first

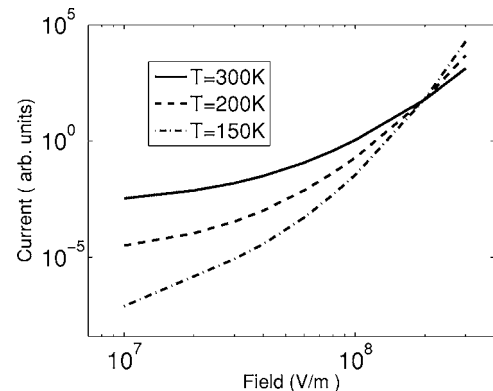


FIG. 3. Temperature dependencies of injection current.

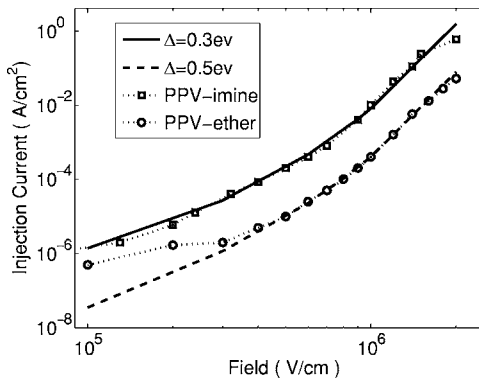


FIG. 4. Comparison between our model and experimental data.

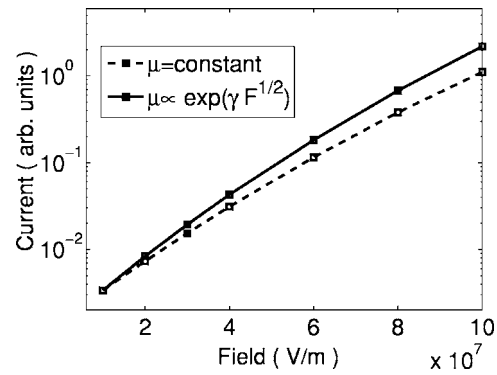


FIG. 5. Comparison between injection currents for field dependent mobility and constant mobility.

substitute Eq. (10) into Eq. (2) to obtain the carrier concentration,

$$p_e(x) = \left[N - \frac{J}{k_B T \mu_0 \exp(\gamma \sqrt{F})} \int_0^x \exp\left(\frac{e\varphi(x')}{k_B T}\right) dx' \right] \times \exp\left(-\frac{e\varphi(x)}{k_B T}\right). \quad (11)$$

Then, by connecting Eqs. (7)–(9) and (11), we obtain the injection current with the field-dependent mobility. Figure 5 illustrates the relation between injection current and electric field with field dependent mobility, with $\mu_0 = 7.3 \times 10^{-6} \text{ cm}^2/\text{V s}$ and $\gamma = 1 \times 10^{-4} \text{ m/V}^{1/2}$, and $\Delta = 0.3 \text{ eV}$. For comparison, the injection current with constant mobility is plotted as well.

The diffusion-controlled injection process in OLED is investigated and the concept of critical distance is proposed. A compact injection model applicable to OLED is formulated. This model is shown to fit the experimental data well and to explain the dependency of the injection current on the barrier height, the temperature, and the electric field. It was found that the field-dependent mobility plays an important role in the injection model at higher electric field.

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