Current Density Equations Representing the Transition between the Injection- and Bulk-limited Currents for Organic Semiconductors

Sang-Gun Lee[a] and Reiji Hattori[b]

Abstract

The theoretical current density equations for organic semiconductors was derived according to the internal carrier emission equation based on the diffusion model at the Schottky barrier contact and the mobility equation based on the field dependence model, the so-called "Poole-Frenkel mobility model." The electric field becomes constant because of the absence of a space charge effect in the case of a higher injection barrier height and a lower sample thickness, but there is distribution in the electric field because of the space charge effect in the case of a lower injection barrier height and a higher sample thickness. The transition between the injection- and bulk-limited currents was presented according to the Schottky barrier height and the sample thickness change.

Keywords: organic semiconductor, theoretical equation, device simulation, injection-limited current, bulk-limited current, transition, ohmic contact

1. Introduction

Compared with inorganic semiconductors, the carrier transportation in organic semiconductors is not yet very clear and is still being debated. Therefore, the current density-voltage (J-V) equation has not been presented clearly in textbooks or research papers. Many organic-semiconductor researchers demand accurate, universal, and theoretical J-V equations. There are two current-limiting processes in organic semiconductors: injection and bulk limitation. The currents that are limited by such processes are called "injection-limited current" (J_{ILC}) and "bulk-limited current" (J_{BLC}), respectively. These two processes coexist under the usual conditions, and the ratio at which the process dominates changes depending on various kinds of conditions, such as the applied voltage, contact barrier height, and sample thickness. This fact makes the equation complicated and prevents the formulation of a standard equation.

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In the case of injection limitation, the Schottky barrier between the metal and organic layers limits the current. Carrier injection into an organic semiconductor is usually represented by Richardson-Schottky thermionic emission. This mechanism is acceptable, however, only for materials with high mobility and a long mean-free path. As organic materials usually have a narrow band and a short mean-free path, the electrons emitted from the metal are immediately scattered very close to the metal and must diffuse inside up to the potential maximum formed by the Schottky and Coulombic potentials against the electric field. This mechanism was discussed in detail for the carrier injection into an insulator [1], and it was concluded that the injection-limited current in insulators has the field dependence of $E^{3/4}\exp(aE^{1/2})$ in a high-field regime. In the case of inorganic semiconductors with low mobility, the carrier injection must be considered based on the carrier diffusion model [2]. This model provides the injection current with a field dependence of $E\exp(aE^{1/2})$ considering Schottky barrier lowering. The similarity of these field dependences stems from the same assumption of physical mechanism. They assume both carrier diffusion and barrier lowering. It is thus no wonder that the diffusion model in inorganic semiconductors has been adapted to organic semiconductors.

On the other hand, in the case of bulk limitation, the injected current makes the space charge in the bulk. In the case of a perfect insulator without traps and field-independent mobility, the bulk limitation current obeys the Mott-Gurney equation [3]. In the case including trap distribution, the so-called "trap-charge-limited current" (TCLC), the equation is given by the reference [4]. These mechanisms, however, are acceptable only for the case of a constant mobility in the organic layer. Furthermore, an approximate analytical solution has been presented by Murgatroyd [5] using Poole-Frenkel field-dependent mobility. The coefficients that were used in their approximated equation, however, were appropriate only for the fixed thickness of their sample. The coefficients must be changed depending on the sample thickness and order of applied voltage. The modified approximate equation, which can follow the sample thickness change, was presented herein.

In carrier transportation in an organic semiconductor, it is generally believed that the current is limited by the injection when the barrier height at the metal/organic interface is more than 0.3~0.4 eV, and that the current is limited by the space charge when the barrier height is less than 0.3~0.4 eV, [6-7]. In this paper, the critical barrier height was fixed at 0.3~0.4 eV, which represented the transition between J_{ILC} and J_{BLC}. In fact, however, they would transit each other when the device parameters (e.g., applied voltage, device thickness, and density of states) change. Therefore, it is demanded that the critical barrier height be expressed by the equation that includes these parameters, which is useful for the device design of OLED.

In this paper, two theoretical equations for organic semiconductors are presented: J_{ILC} and J_{BLC}. The current density, J_{ILC}, is based on the diffusion theory [2], which includes the parameters not only of the injection barrier height but also of mobility and the effective density of states, which means that J_{ILC} cannot be determined without these parameters. On the other hand, J_{BLC} is based on the field-dependent carrier mobility. When the space charge effect is considered, the electric field at the injection interface between the metal and organic layers is usually assumed to be zero. A zero electric field cannot generate the injection current, however, and a small electric field at the injection interface must be considered even in the case where J_{BLC} is dominant, which means that these effects must be considered simultaneously. In this case, however, the *J-V* equation can no longer be derived analytically; as such, the J_{ILC} and J_{BLC} equations were derived separately and were combined using the simple equation, which is convenient and useful for analysing the experiment results. In addition, assuming the condition of J_{ILC}=J_{BLC}, the critical barrier height, which pertains to how low the barrier height should be to obtain ohmic contact, can be obtained.

There are two ways of confirming the accuracy of the theoretical *J-V* equations. The general method involves comparing them with experiment results. In this method, the parameters of the sample, such as the layer thickness, barrier height, or mobility, have to be changed. Almost all the parameters have to be changed during the fabrication process, however, for which reason the samples have to be fabricated one by one while changing the conditions. In this case, it is hard to keep the same fabrication conditions. In addition, it is also difficult to change the parameters individually because the fabrication parameters are closely related to one another. The other method involves comparing the theoretical *J-V* equations for organic semiconductors with the results of the device simulation, where it is easy to change the individual parameters. Therefore, the validity of the theoretical *J-V* equations for organic semiconductors was confirmed by comparing them with the results of the device simulation rather than with the experiment results.

The objective of this work was to derive the theoretical *J-V* equations that appear in a textbook of organic electronics. In the previous work [8], it was assumed that an electric field becomes constant because of the absence of a space charge effect, but it was found that the field distribution had to be considered in the case of a low barrier height. Therefore, in this work, the theoretical equations were extended from the no-space-charge effect to the space charge effect according to the Schottky barrier height and sample thickness change, and the transition between the injection- and bulk-limited currents was presented.

2. Device Structure and Material Parameters

Fig. 1 shows the energy diagram of organic semiconductors. When the barrier height is sufficiently high (a), the electric field becomes constant because of the absence of a space charge effect. On the other hand, when the barrier height is sufficiently low (b), the injected current forms a space charge in the bulk, which limits the current because it decreases the electric field.

How each parameter affects the *J-V* characteristics was evaluated using the 2D device simulator (ATLAS Sil-

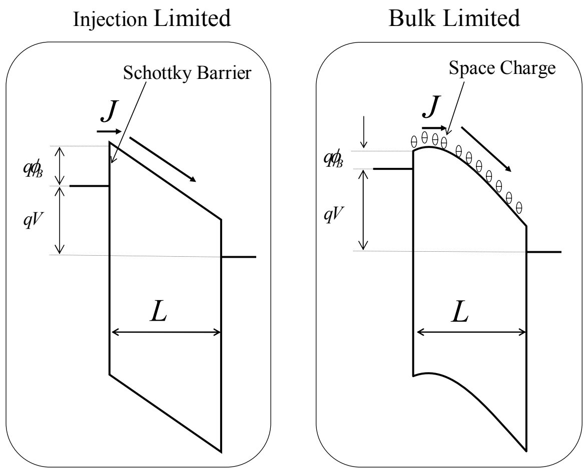

Fig. 1. Schematic energy band diagram for organic semiconductors.

Table 1. Material Parameters Used for the Device Simulation.

Parameters	Organic Layer	Unit
Barrier height (ϕ_B)	0.01~0.5	eV
Band gap (E_g)	2.6	eV
Electric affinity (χ)	3.1	eV
Prefactor mobility (μ_0)	5.00E-06	$cm^2V^{-1}s^{-1}$
Activation energy (ε_a)	0.48	eV
PF factor (β)	3.70E-04	$V^{-1/2}cm^{-1/2}$
Barrier-lowering factor (γ)	0	$V^{-1/2}cm^{-1/2}$
Relative dielectric constant (ε)	3.1	
Effective DOS (N_C)	2.50E+ 19	cm^{-3}
Sample thickness (L)	25~800	nm

vaco Co.). Although this device simulator is generally used for silicon or compound semiconductor devices, it can be applied to organic semiconductors according to the internal carrier emission equation based on the diffusion model at the Schottky barrier contact and the mobility equation based on the field dependence model. Table 1 shows the physical-parameter values that were used in the device simulation, which were derived from the related literature.

3. Derivation of the J-V Equations for Organic Semiconductors

3.1 Injection-limited current (J_{ILC})

Generally, the thermionic-emission model has been used to represent the injection current from the metal to the organic material through the Schottky barrier. This mechanism, however, is appropriate only in semiconductors with high mobility and a long mean-free path. In the case of organic semiconductors with low mobility and a short mean-free path, the injection current should follow a diffusion model, where the emitted carriers are scattered immediately and accumulated near the interface, so that the carriers diffuse into the direction opposite that of the carrier flow. In this case, the current flow is related to how much room exists in the density of states. As such, the injection current is proportional to the mobility, density of states, and electric field near the metal. Therefore, the injection current must use the diffusion model in organic materials because of the low mobility therein, which is given as follows:

$$J_{ILC} = q\mu EN \exp\left(-\frac{q\phi_B}{kT}\right)\exp\left(\gamma\sqrt{E}\right), \tag{1}$$

where q is an elemental charge, μ the mobility, E the electric field, N the effective density of states, and ϕ_B the injection barrier height. The barrier lowering due to the image force is expressed by the last term, with the factor γ [9].

The mobility in organic semiconductors is often analyzed using either the modified Poole-Frenkel model (PFM) or the Gaussian disorder model (GDM). PFM can be described as a carrier from the coulomb potential of a charged trap [10], given as follows:

$$\mu_{PF} = \mu_{PF0} \exp\left(-\frac{\varepsilon_a}{kT}\right)\exp\left(\beta\sqrt{E}\right), \tag{2}$$

where μ_{PF0} is the temperature-independent prefactor mobility, ε_a the thermal-activation energy of the trapped carrier, and β the Poole-Frenkel factor as a fitting parameter.

Another model for field-dependent mobility is GDM by Bässler [11]. GDM can be described as a biased random walk among dopant molecules with Gaussian-distributed random-site energies [12], given as follows:

$$\mu_{GM} = \mu_{GM0} \exp\left\{-\left(\frac{2\sigma}{3kT}\right)^2\right\}\exp\left[C\left\{\left(\frac{\sigma}{kT}\right)^2 - \Sigma^2\right\}\sqrt{E}\right], \tag{3}$$

where μ_{GM0} is the temperature-independent prefactor mobility, σ the width of the energetic disorder, C the potential-disorder parameter, and Σ an empirical constant.

Equations (2) and (3) are based on totally different physics and have different temperature dependences but have the same field dependence and can be expressed in the same equation, as follows:

$$\mu = \mu_0 \exp\left(\eta\sqrt{E}\right), \tag{4}$$

By substituting this into equation (1) and assuming $E=V/L$ over the whole layer, the following equation can be obtained:

$$J_{ILC} = q\mu_0 N \frac{V}{L} \exp\left(-\frac{q\phi_B}{kT}\right) \exp\left\{(\eta+\gamma)\sqrt{\frac{V}{L}}\right\}, \tag{5}$$

This is the *J-V* equation for the injection-limited current (J_{ILC}).

3.2 Bulk-limited current (J_{BLC})

Fig. 2 shows the internal electric-field distributions as a parameter of injection barrier height at the 10 V applied voltage, calculated via device simulation. In the case of a barrier height of more than 0.2 eV, the electric field becomes constant over the whole layer as the adequate amount of space charge is not yet generated. In the case of a barrier height lower than 0.1 eV, however, the electric field can no longer be assumed to be constant. Therefore, the theoretical *J-V* characteristics must include the space charge effect in the case of a low injection barrier height.

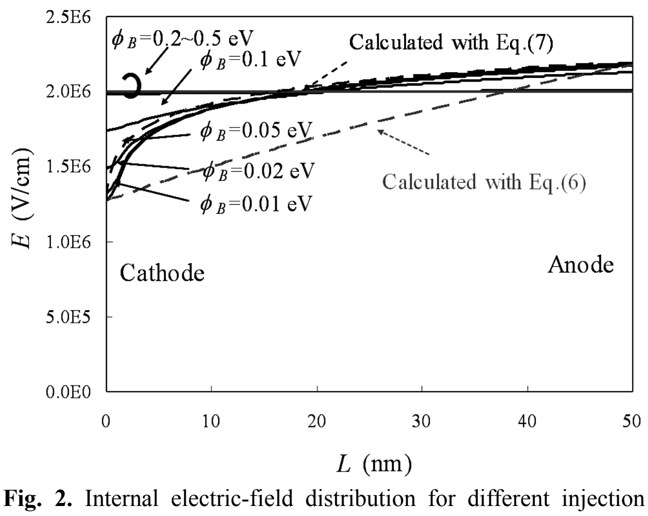

Fig. 2. Internal electric-field distribution for different injection barrier heights at the 10 V applied voltage.

The equation for electric-field distribution, including the space charge effect, should be solved using the differential equation according to $dE/dx = -qn/\varepsilon_r\varepsilon_0$ (Poisson's equation) and $J = -qn\mu E$ (continuity equation). If the mobility is independent of the electric field, this equation can be easily solved and is well known as Mott-Gurney's equation, as follows:

$$E = \sqrt{\left(2J_{BLC}/\mu\varepsilon_r\varepsilon_0\right)x + E_0^{\ 2}}, \tag{6}$$

where ε_r is the relative permittivity, ε_0 the vacuum permittivity, and E_0 the electric field at x=0 as an initial condition. The dotted gray line in Fig. 2 is calculated using equation (6), which differs largely from the device simulation. The reason for this is that the mobility is assumed to be constant. On the other hand, in the case of the mobility that has field dependence, as shown in equation (4), the following equation is obtained [5, 13]:

$$\frac{2\mu_0\varepsilon_r\varepsilon_0}{\eta^4 J_{BLC}}\left(\eta^3 E^{3/2} - 3\eta^2 E + 6\eta\sqrt{E} - 6\right)\exp\left(\eta\sqrt{E}\right) = x + x_0, \tag{7}$$

where the electric field, E, is varied with x, and x_0 is decided based on E_0 as an initial condition, as follows:

$$x_0 = \frac{2\mu_0\varepsilon_r\varepsilon_0}{\eta^4 J_{BLC}}\left(\eta^3 E_0^{\ 3/2} - 3\eta^2 E_0 + 6\eta\sqrt{E_0} - 6\right)\exp\left(\eta\sqrt{E_0}\right), \tag{8}$$

The dotted black line in Fig. 2 is calculated using equation (7), which can have relatively good agreement with the results of the device simulation. To obtain the *J-V* relation, equation (7) must again be integrated with x, but the equation then becomes too complicated to deal with. Therefore, if E is assumed to be very large under the condition at $x=L$, E_0 and x_0 can be neglected, and the approximate expression can be obtained as follows:

$$J_{BLC} \approx \frac{2\mu_0\varepsilon_r\varepsilon_0}{\eta}\alpha^{3/2}\frac{V^{3/2}}{L^{5/2}}\exp\left(\eta\sqrt{\alpha\frac{V}{L}}\right), \tag{9}$$

where $E=\alpha(V/L)$ is substituted into equation (7), and α is an adjusting parameter to fit E to the real value. This is the *J-V* equation for the bulk-limited current (J_{BLC}) with field-dependent mobility.

3.3 Transition between J_{ILC} and J_{BLC}

The theoretical *J-V* equations (5) and (9), which are valid at different conditions (i.e., J_{ILC} and J_{BLC}, respectively), were obtained. The *J-V* characteristics, however, must be considered J_{ILC} and J_{BLC} simultaneously. One of the simplest ways to do this is to make the total current density follow equation (10) below.

$$J_{Total} = \frac{J_{ILC} \cdot J_{BLC}}{J_{ILC} + J_{BLC}}, \tag{10}$$

This equation shows that the total current is simply limited by the smaller current, J_{ILC} or J_{BLC}. This equation is not derived from an accurate analysis, but it well expresses the transition between J_{ILC} and J_{BLC}.

Fig. 3 shows the *J-V* characteristics as a parameter of injection barrier height obtained via device simulation and the theoretical equation, using equation (10). The sample thickness was fixed at 50 nm. In both calculations, the same physical model and parameters were employed. As shown in Fig. 3, both curves completely overlap, which means that the theoretical equation can express both currents with a simple equation. It was found that J_{BLC} is lower than in the case of ϕ_B=0.1 eV, and that J_{ILC} is higher than in the case of ϕ_B=0.2 eV. Therefore, the current equation can be successfully derived as a parameter of injection barrier height.

Fig. 4 shows the *J-V* characteristics as a parameter of sample thickness obtained via device simulation and the theoretical equation, using equation (10). To enhance the effect of J_{BLC}, the ϕ_B was fixed at the sufficiently low level of 0.1 eV. In both calculations, the same physical model and parameters were employed. As shown in Fig. 4, relatively good agreement between the two sets of results was obtained, showing the transition between J_{ILC} and J_{BLC}, expect for cases with higher sample thickness. The deviations in sample thickness are due to the approximation in equation (7), where the electric field must be sufficiently high.

Fig. 5 shows the critical barrier height (ϕ_{Bc}) dependence on sample thickness at the 10 V applied voltage. Assuming the condition of $J_{ILC}= J_{BLC}$, ϕ_{Bc} can be obtained, as shown in equation (11), which pertains to how low the barrier height should be to obtain ohmic contact. In other words, the barrier height (ϕ_B) does not have to be less than ϕ_{Bc}. This figure indicates that a thicker sample needs a lower barrier height to obtain ohmic contact. This is because the space charge effect easily occurs when the sample is thicker. Notice that mobility does not appear in this equa-

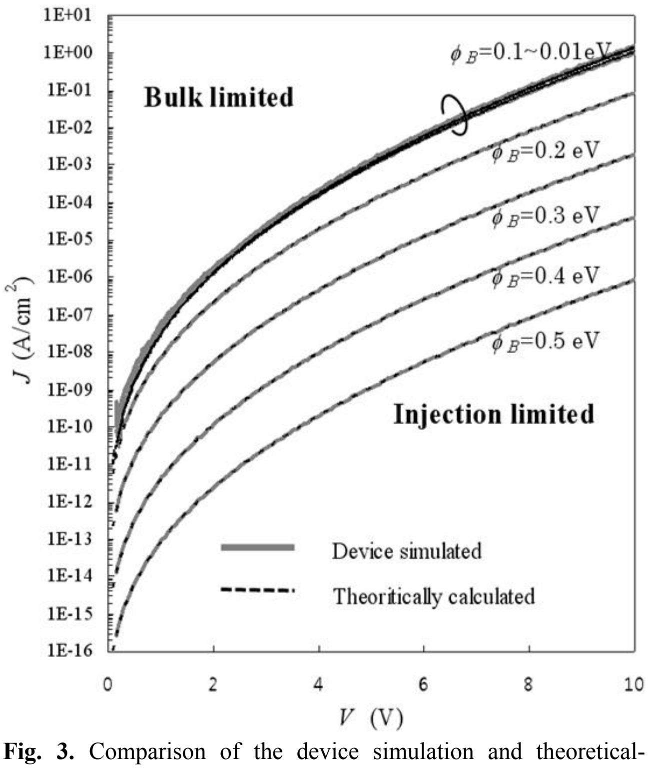

Fig. 3. Comparison of the device simulation and theoretical-calculation results as parameters of injection barrier height.

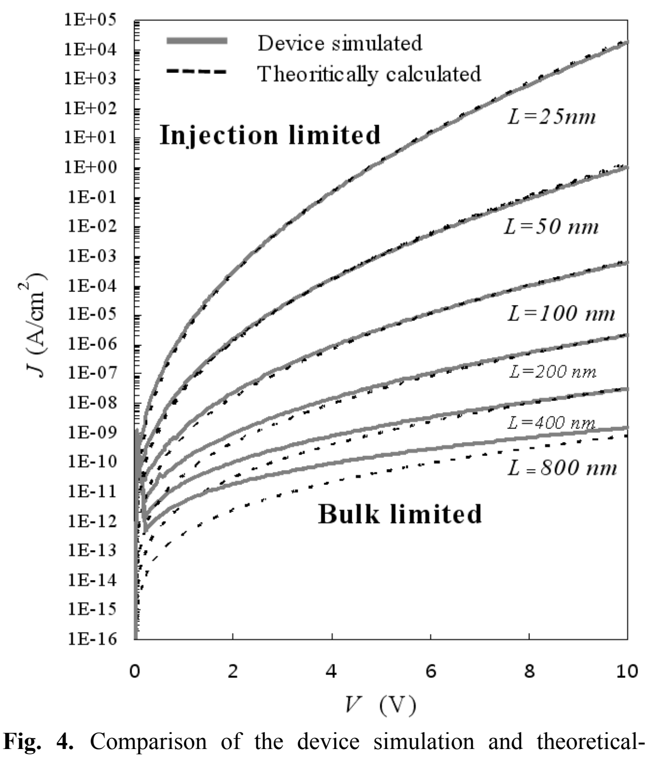

Fig. 4. Comparison of the device simulation and theoretical-calculation results as parameters of sample thickness.

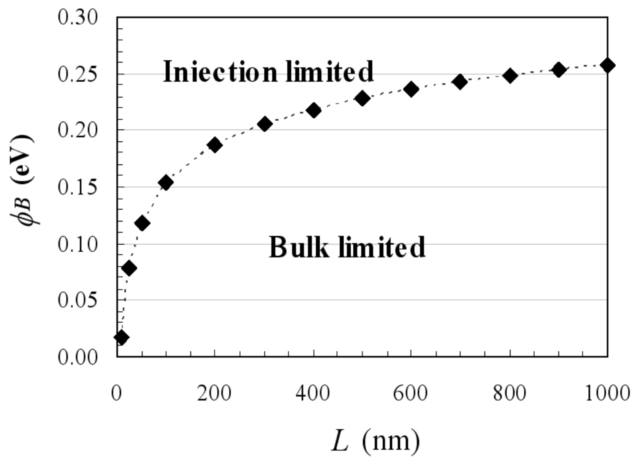

Fig. 5. Sample thickness dependence of the critical barrier height separating J_{ILC} and J_{BLC}.

tion, which means that it has no effect on the contact characteristics. ϕ_{Bc}, however, expressly depends on the sample thickness and the density of states in the device parameter. Therefore, $\boldsymbol{\phi_{Bc}}$ is transited by the device parameters, as shown in equation (11).

$$\phi_{Bc} = \frac{kT}{q}\left[\sqrt{\frac{V}{L}}\left\{\eta\left(1-\sqrt{\alpha}\right)+\gamma\right\}-\ln\left(\frac{2\varepsilon\alpha^{3/2}}{qNL\eta}\sqrt{\frac{V}{L}}\right)\right], \qquad (11)$$

Therefore, the transition of the *J-V* characteristics between J_{ILC} and J_{BLC} can be known, as shown in Fig. 5. For example, in the case of sample thickness (L=50 nm), an ϕ_B that is less than 0.12 eV has no effect on the current. In other words, an ϕ_B that is less than 0.12 eV can consider ohmic contact for organic semiconductors.

4. Conclusions

The theoretical equations of the J-V characteristics for organic semiconductors were successfully derived based on the diffusion and field dependence mobility models. The internal electric field becomes constant over the whole layer in the case of a higher injection barrier height and a lower sample thickness, so that the theoretical equations must consider only J_{ILC}. As the internal electric field has distribution, however, in the case of a lower injection barrier height and a higher sample thickness, the theoretical equations must consider J_{BLC}. Therefore, the accurate theoretical equations for organic semiconductors must simultaneously consider J_{ILC} and J_{BLC}.

Assuming the condition of J_{ILC}=J_{BLC}, the critical barrier height, which pertains to how low the barrier height should be to obtain ohmic contact, can be obtained. Thus, the aforementioned equation can successfully represent the transition between J_{ILC} and J_{BLC} when the device parameters (e.g., applied voltage, device thickness, and density of states) change. The transition of the *J-V* characteristics can help in understanding the electrical-transport mechanism of organic semiconductors and will be very useful in explaining various experiment results.

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