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Electrical model of organic diodes with field-dependent carrier mobility in the presence of an electric field at the injection interface

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SUMMARY

In this study, we propose a model to simulate electrical conduction of single carrier organic diodes in the presence of a barrier for carrier injection and considering a field-dependent carrier mobility. An analytical expression for the internal electric field function, that simplifies operation with the resulting nonlinear equation system, is provided. Simulated results with this model are compared with those obtained from other existing approaches that assume simplifications. Current density versus voltage curves predicted by this model for diodes with different active layer thicknesses show good agreement with the experimental results. Copyright © 2010 John Wiley & Sons, Ltd.

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KEY WORDS: OLED; PLED; interface field; mobility

1. INTRODUCTION

Modeling of the electrical behavior (current density versus voltage, J–V) of organic light-emitting diodes (OLEDs) is of crucial interest to design appropriate OLED display drivers. One of the main difficulties found when modeling J–Vs of OLEDs is to determine the internal electric field along the active layer under external biasing (V). This problem arises from the space charge effect due to the excess of injected carriers, which contribute to this field. The space charge distribution is, in turn, a nonlinear function of the spatial coordinate due to the low carrier mobilities in organic materials. Moreover, one should take into account the virtual dependence of carrier mobility on both charge density [1] and electric field [2]. Difficulties become very hard

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to overcome when multilayer structures are used, and both carrier types, electrons and holes, are involved in conduction. In general, solution-processed devices reduce the number of layers to three or less, due to incompatibility with solvents, but despite simplifications of the diode structure, and even working with single-carrier devices, mathematical modeling usually resort to approximations. In the limit case with devices involving high barriers for carrier injection (>0.3 eV), current is so low that the internal electric field (E) resembles that of a capacitor, and it may be assumed $E = V/L$, where L is the active layer thickness. In the other limit case, when there is no injection barrier (ohmic contact), the electric field is totally screened at the interface and eventually shows a spatial dependence of the square-root type, as expected from a pure space charge effect. Actually, most situations fall within both the extremes, as it is difficult to avoid a small barrier for carrier injection.

In this study, we propose a numerical conduction model for single-carrier organic diodes that solves the electric field inside the sample, in the presence of a barrier for carrier injection and assuming a field-dependent carrier mobility for bulk transport. This model simulates J–V curves using a microscopic approach for carrier injection based on a microscopic hopping theory [3]. Results are compared with previous empirical models [2], and more recent approaches [4], and are used to predict J–V characteristics for samples with different active layer thicknesses.

2. THEORETICAL MODEL

The proposed model is based on the numerical determination of the electric field at both interfaces, $E_0 = E(x = 0)$ and $E_L = E(x = L)$, between metal contacts and organic material. A field-dependent mobility is considered,

$$\mu(E) = \mu_0 \exp(k\sqrt{E}) \tag{1}$$

where μ_0 is the mobility at zero field and k the phenomenological factor. This expression is included in the drift current, which combined with the Poisson equation is integrated to express the electric field function $E(x, E_0, J)$ along the spatial coordinate x, where J is the current density:

$$x = \frac{\phi\varepsilon\mu_0}{J}\frac{2}{k^4}[F(E) - F(E_0)] \tag{2}$$

where $F(E) = e^{k\sqrt{E}}(k^3E^{3/2} - 3k^2E + 6kE^{1/2} - 6)$, ε is the material dielectric constant and ϕ is the ratio between the carrier density contributing to the transport and the total injected charge [5]. Evaluation of both members at $x = L$ provides a first nonlinear equation $f(E_0, E_L, J) = 0$.

Further integration in the x coordinate of the electric field between its two extreme values (E_0 and E_L) allows to obtain the external applied voltage, providing a second nonlinear equation $g(E_0, E_L, V, J) = 0$,

$$V = \frac{\phi\varepsilon\mu_0}{J}\frac{2}{k^6}[G(E_L) - G(E_0)] \tag{3}$$

where $G(E) = e^{k\sqrt{E_L}}(k^5E_L^{5/2} - 5k^4E_L^2 + 20k^3E_L^{3/2} - 60k^2E_L + 120kE_L^{1/2} - 120)$.

The continuity equation for the current density at the interface, under steady regime (in the absence of carrier recombination), provides a third nonlinear equation:

$$J_{\text{inj}}(E_0) = J(V, E_0, E_L) \tag{4}$$

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where J_{inj} is the injection current. Equation (4) is a general expression valid for any injection model proposed in the literature. Notice that carrier injection is always dependent on the electric field at the interface (E_0). In this study, J_{inj} is given by the microscopic hopping model proposed by Arkhipov *et al.* [3]. Finally, a subroutine for nonlinear equation systems was used to solve, for each bias V, the extreme values E_0 (electric field at the anode) and E_L (electric field at the cathode) from Equation (3), and (2) evaluated at $x = L$. For this purpose, a FORTRAN recipe that solves nonlinear equation systems using a modified Powell hybrid algorithm and a finite-difference approximation to the Jacobian was chosen. Once E_L and E_0 values were determined, J is evaluated, delivering the J–V characteristic.

3. ANALYTICAL APPROACH FOR THE ELECTRIC FIELD

The previous routine gives excellent results for most of the physical cases considered, although the computing time is large, and occasional problems with the algorithm convergence were observed in the range of low voltages, depending on the initial guess for E_0 and E_L.

A simplification of the previous system is provided by means of a Taylor expansion of the spatial coordinate x given by (2), in power series of the field E around the interface value E_0:

$$x = K[A(E - E_0)^3 + B(E - E_0)^2 + C(E - E_0)] \tag{5}$$

where the independent term vanishes because $x = 0$ when $E = E_0$, and the coefficients result:

$$K = \frac{\phi\varepsilon\mu_0}{J} \tag{6}$$

$$A = e^{k\sqrt{E_0}}\frac{k}{24}\left(k + \frac{3}{\sqrt{E_0}}\right) \tag{7}$$

$$B = \frac{e^{k\sqrt{E_0}}}{4}(k\sqrt{E_0} + 2) \tag{8}$$

$$C = e^{k\sqrt{E_0}}E_0 \tag{9}$$

Notice that for $k = 0$, the third order term vanishes, and the resulting expression equals the analytic one obtained for a constant mobility [6]:

$$x = \frac{\phi\varepsilon\mu_0}{2J}(E^2 - E_0^2) \tag{10}$$

It has been checked that a third order expansion is good enough to guarantee an error in the spatial coordinate $<2\%$ for typical $k < 10^{-3}$ $(\mathrm{m/V})^{1/2}$, with respect to the exact numerical solution.

Under this approach, the electric field function $E(x)$ in (5) may be analytically solved. This is of great help to obtain $E_L = E(L, E_0)$, and consequently, to express the external voltage as $V(J_{inj}, E_L, E_0)$ in (3). Thus, for a given V, E_0 may be determined from (3). For that purpose, we have used a simple FORTRAN routine to find zeros using the Müller's method. Subsequently, the current density $J(E_0)$ is determined and then the J–V characteristic.

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4. RESULTS

Figure 1 shows the exact electric field distribution $E(x)$, as obtained from (2) after solving the nonlinear equation system, for different injection barriers (a) and k factors of the mobility function (b). A zero-field mobility $\mu_0 = 7.7 \times 10^{-7}\,\mathrm{m^2/V\,s}$, and an active layer thickness of 100 nm were chosen as material characteristics. The external applied voltage was set to 20 V. For those conditions, the current density lowers with increasing injection barrier as expected. In this case, the computed currents were 495, 372 and 158 $\mathrm{A/m^2}$, for the barriers 0.4, 0.5 and 0.6 eV displayed in Figure 1(a), respectively. As explained in Section 1, as current density reduces, the E_0 grows and approaches V/L, like in a capacitor. When E_0 is computed for a constant current density with different injection barriers, it is observed that E_0 lowers with reducing the barrier. This is due to the fact that the external voltage must also be significantly decreased to keep a constant current when reducing injection barrier.

On the contrary, Figure 1(b) shows that E_0 grows toward the V/L value when the k factor and eventually the mobility are increased. A reduction in mobility means an accumulation of charge in the active layer, which results in a higher space-charge effect and a stronger screening of the electric field at the interface. In contrast, when the mobility is high, like in typical inorganic semiconductors, injected carriers are quickly evacuated through the contacts. Thus, the accumulated space charge is residual, and the electric field across the active layer approaches the constant value V/L. In all the cases, it was found that the field distribution clearly differs from the square root law typically obtained under pure space charge regime.

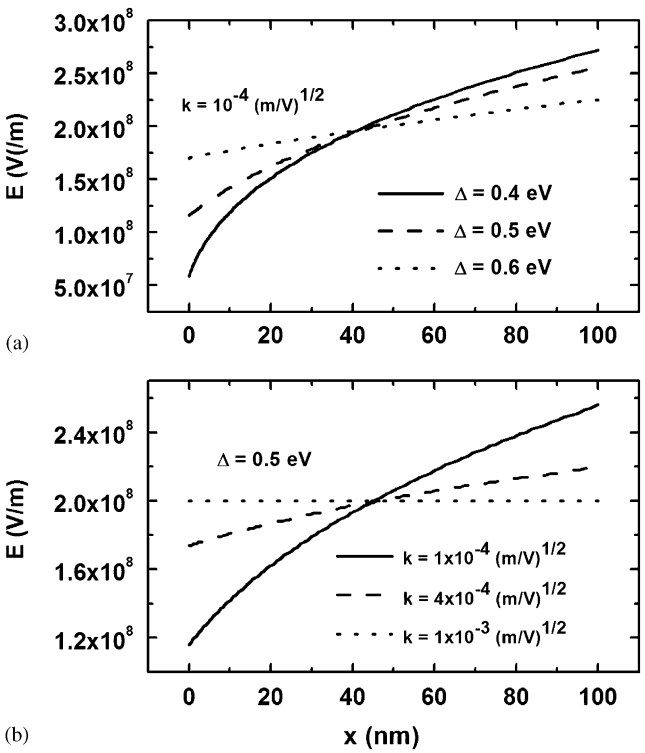

Figure 1. Simulation of the electric field (E) distribution along the vertical coordinate x (nm), in diodes with different (a) injection barriers (Δ), and (b) k factors (mobilities).

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Figure 2 shows different J–V characteristics calculated using this model (open symbols). These are compared with the approximation of Murgatroyd (solid symbols) [2], who assumes a vanishing electric field at the interface.

It is concluded that for those systems with a significant carrier injection barrier, such as most diodes fabricated using ITO/PEDOT at the hole injection electrode, theoretical approaches that do not consider the existence of the electric field at the interface may fail already at low voltage values.

The result of applying our model to study the polymer-type OLEDs (PLEDs) electrical behavior is presented in Figure 3. This figure shows the experimental J_{exp}–V_{exp} of three diodes based on the structure: ITO/PEDOT:PSS/PFP:$(CN)_2$/Al, where PFP:$(CN)_2$ refers to a polyfluorene derivative [7].

The model proposed in the earlier section was fitted to a test experimental J_{exp}–V_{exp}, using the mentioned injection function $J_{inj}(E_0)$ proposed by Arkhipov *et al*. [3]. A number of physical parameters are involved in this model: barrier height for carrier injection (Δ), energetic width (σ) of the density of states for the energy level under Gaussian approximation, carrier inverse localization radius (γ), average nearest neighbor-hoping distance (a), and the attempt-to-jump frequency (v_o). All of them are well described in [3], and most of them may be physically measured, although not with similar accuracy.

The fitting procedure has been performed using a non-deterministic genetic algorithm, as this type of algorithms is considered more suitable for systems with many parameters [8]. In our case, these parameters are: μ_0, k, Δ, γ, and a. Attempt-to-jump frequency, v_o, was previously obtained for each PLED from their electroluminescence (EL) spectra at high bias. As it is assumed that v_o is enhanced by lattice vibrations, spectrally resolved vibronic features allow a good estimation of this parameter, resulting $v_{o(PFP:CN2)} = 4.75 \times 10^{13}$/s. σ was approximated by Gaussian deconvolution of the excitonic emission in the EL spectrum, obtaining: $\sigma = 63.4$ meV.

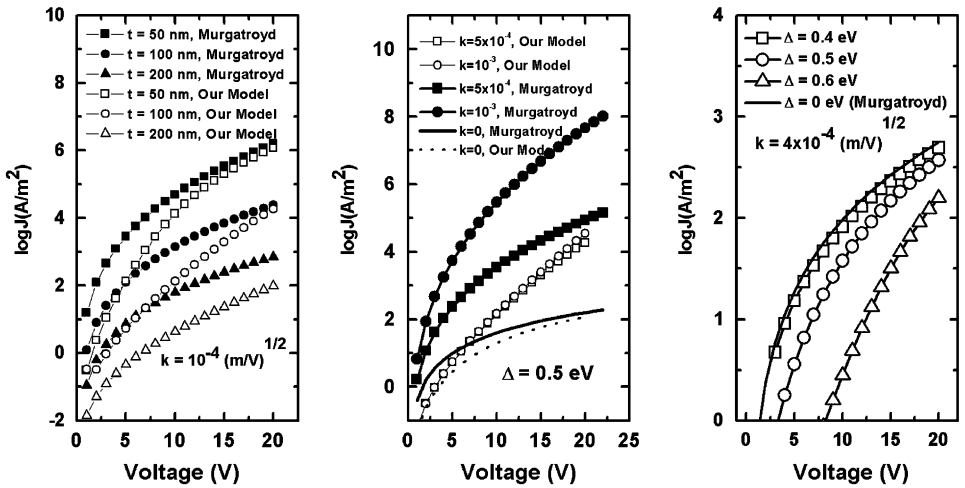

Figure 2. Comparison between the current density–voltage (J–V) characteristics obtained with the proposed model (open symbols) and those obtained by the approach of Murgatroyd (solid symbols) for: (a) different thicknesses (same injection barrier, D, and mobility), (b) different k factors (same thickness and injection barrier), and (c) different barriers (same thickness and mobility).

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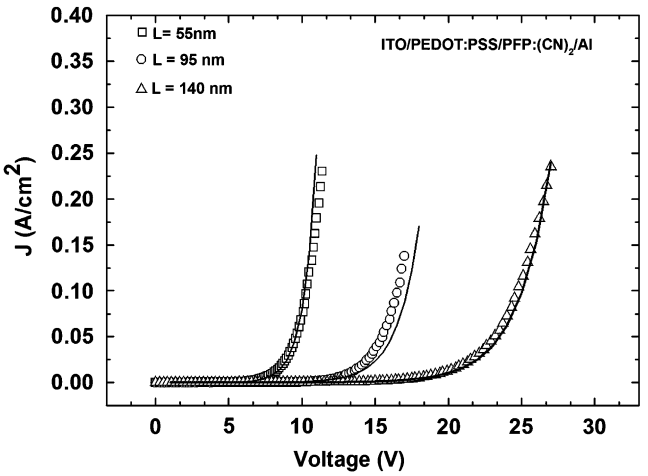

Figure 3. Experimental (symbols) and theoretical current density versus voltage curves (solid lines) for diodes with different active layer thicknesses: 55 nm (squares), 95 nm (circles), and 140 nm (triangles).

Parameters for the PFP:$(CN)_2$-based PLED are: $\Delta = 6.7\,\text{eV}$, $a = 1.05\,\text{nm}$, $\gamma = 3.8 \times 10^9/\text{m}$, $k = 7.9 \times 10^{-4}\ (\text{m/V})^{1/2}$ and zero-field hole mobility $\mu_0 = 5.5 \times 10^{-7}\,\text{m}^2/\text{V s}$.

To demonstrate the validity of this model, diodes of the same material but different active layer thicknesses (ranging from 55 to 140 nm) were fabricated and characterized (Figure 3). Active layer thicknesses were determined by profilometry. The model described above was previously fitted to one sample, obtaining a set of parameters. Next, this set of parameters was used to simulate *J–V* curves for devices with the corresponding active layer thicknesses. Experimental *J–V* curves (dotted lines) and theoretical simulations (solid lines) are plotted in Figure 3. Assuming measurement uncertainties and disregarding non-ideal behavior such as leakage currents, the agreement between theory and experiment is good.

5. CONCLUSIONS

A complete model to simulate electrical conduction in single carrier organic diodes has been developed. This model takes into account a non-negligible carrier injection barrier (non-vanishing electric at the interface) and a field-dependent mobility, as expected for systems working at low carrier densities.

A numerical solution for the internal electric field is calculated for several operating conditions and compared with results of other existing models.

To simplify calculations, which require time-consuming nonlinear system subroutines, an approach consisting of a Taylor expansion up to third order in the electric field of the spatial coordinate is derived and proved to be accurate within an error $<2\%$.

The model has been fitted to experimental *J–V* curves to obtain the material and structural parameters that control carrier conduction. Using these general parameters, predictions of this model are in good agreement with the experimental *J–V* curves of devices with different active layer thicknesses.

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