Thickness scaling of the space-charge-limited current in poly(p-phenylene vinylene)
Blom, P. W. M.; Tanase, C.; de Leeuw, D. M.; Coehoorn, R.

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Charge transport in light-emitting diodes (LEDs) based on a poly(p-phenylene vinylene) (PPV) derivative is investigated as a function of sample thickness. Via the thickness dependence, the contributions from the electric field and charge carrier density to the mobility in space-charge-limited (SCL) diodes can be disentangled. It is demonstrated that a field-dependent mobility weakens the thickness dependence of the SCL current, whereas a carrier-density-dependent mobility gives rise to an enhanced thickness dependence. The enhanced thickness dependence of the experimental SCL current in PPV is in agreement with the predictions using a density-dependent mobility only. This observation confirms that in PPV-based LEDs, the hole transport is dominated by filling of the localized states. © 2005 American Institute of Physics. [DOI: 10.1063/1.1868865]
of the ITO, a 100–1000 nm polymer film has been spin coated from toluene solution. The device was finished by thermal evaporation of silver (Ag) through a shadow mask. The hole-only diodes have been measured under a controlled N₂ atmosphere. The electrical measurements have been performed using a Keithley 2400 SourceMeter.

In Fig. 1 the current density-voltage (J-V) measurements are presented for NRS-PPV hole-only diodes with thicknesses L of 200, 560, and 950 nm. The applied voltage is corrected for the built-in voltage V₀ of 1 V resulting from the work function difference between ITO and Ag. As a first step, we check whether the experimental current at low voltages obeys the conventional Mott–Gurney (MG) law given by

\[ J_{\text{MG}} = \frac{9}{8} \varepsilon_0 \varepsilon_r \mu_0 (0, T) \frac{V^2}{L^3}, \]

with \( \varepsilon_0 \varepsilon_r \), the dielectric constant, as indicated by the solid lines in Fig. 1. The observed occurrence of a SCL current enables a direct determination of the hole mobility for NRS-PPV at low voltages, which is \( 5.0 \times 10^{-12} \text{ m}^2/\text{V s} \) [Eq. (3)].

For a large field-enhancement factor \( \gamma \) the exponential factor will dominate the increase of the current. As a result, \( V(L) \) curves at a fixed current density fall in between the curves \( V=V_0 (L/L_0) \) and \( V=V_0 (L/L_0)^{3/2} \), depending on the magnitude of \( \gamma \). Here \( V_0 \) is the voltage at a certain reference thickness \( L_0 \), at the chosen fixed current density. In other words, the increase of the voltage with increasing thickness, at a fixed current density, is smaller than would be expected on the basis of Eq. (3). On the other hand, for a density-dependent mobility \( \mu_s(p) \) according to Eq. (2), we have recently demonstrated that it can be approximated by

\[ J = 0.8 \varepsilon_0 \varepsilon_r \mu_0 (p_{sv}) E_{sv}^2, \]

with \( E_{sv} = V/L \), \( p_{sv} \), the average density in the device given by \( p_{sv} = (3/2) (\varepsilon_0 \varepsilon_r V/e L^2) \), and \( \mu_0 (p_{sv}) \) the mobility at density \( p_{sv} \). Combining this equation with Eq. (2) leads to a thickness dependence of the form

\[ J = J_{\text{MG}} + c \frac{V}{L^2} \left( \frac{V}{L^2} \right)^{T_0/T-1} \frac{V^{T_0/T+1}}{L^{2(T_0/T+1)}}, \]

where \( c \) is a proportionality constant. For \( T_0 = T \) this will lead to the conventional \( V^2/L^3 \) behavior. For the materials of interest, \( T_0 \) is well above room temperature. For \( T_0 \gg T \), and enhanced by a field-dependent mobility \( (V/L) \) and enhanced by a density-dependent mobility \( (V/L^2) \). Consequently, the thickness dependence of the SCL current at high voltages can be used to discriminate between the contributions from field and charge carrier density.

In order to exactly model the SCL currents with either \( \mu_0 (E) \) or \( \mu_0 (p) \), Eqs. (1) and (2) are combined with

\[ J = p(x) \varepsilon_0 \varepsilon_r \mu_0 (p(x), E(x)) \frac{dE(x)}{dx} = p(x), \]

with \( p(x) \) the density of holes, and \( E(x) \) the electric field. These equations are solved numerically for a given hole current density \( J \). In Fig. 2 the experimental J-V characteristics are shown together with the numerical model calculations using a field-dependent mobility only. As a reference, the 200 nm device is fitted to determine the field-enhancement factor \( \gamma \) of \( 5 \times 10^{-4} \text{ m/V}^{3/2} \), close to earlier results on...
PPV-based FETs have been investigated. From the temperature dependence, the following parameters for NRS-PPV have been determined: $\sigma_0=3.5 \times 10^8 $ S/m, $\sigma^2=1.36$ Å, and $T_0=560$ K, as obtained from field-effect measurements.

$OC_3C_{10}$PPV. This $\gamma$ value is then used to predict the SCL currents for the thicknesses of 560 and 950 nm devices. As shown in Fig. 2, the predicted currents (solid lines) clearly overestimate the SCL currents at high voltages. Apparently, the calculated thickness dependence using $\mu_c(E)$ is to weak. Fitting the experimental data would lead to $\gamma$ values of $3 \times 10^{-4}$ (m/V)$^{1/2}$ and $2 \times 10^{-4}$ (m/V)$^{1/2}$ for the 560 and 950 nm devices, respectively. A thickness-dependent $\gamma$ is, of course, not physical.

In a recent study, the transfer characteristics of NRS-PPV-based FETs have been investigated. From the temperature dependence, the following parameters for NRS-PPV have been determined: $\sigma_0=3.5 \times 10^8 $ S/m, $\sigma^2=1.36$ Å, and $T_0=560$ K. The numerically calculated SCL currents using the density-dependent mobility with these parameters in Eq. (2) are shown in Fig. 3, together with the experimental data. It appears that the predicted SCL currents using $\mu_c(p)$ are in good agreement with the experimental SCL current for the whole thickness range studied. Note that the calculated currents do not contain any fit parameter. This result demonstrates that the experimental $J-V$ characteristics indeed exhibit the enhanced thickness dependence, as expected from the density-dependent mobility. This observation is therefore a clear proof of the dominance of a density-dependent mobility in the current in polymeric LEDs. The injected charges will first occupy the energetically lowest localized states of the organic semiconductor. With increasing voltage, the additional charges in the SCL diode fill up higher states and therefore will need less activation energy for hops towards neighboring sites. As a result, the charge carrier mobility will be enhanced at higher voltages. The fundamental question as to whether the increase of the mobility in a SCL device is dominated by either the carrier density or the electric field is relevant for the operation of LEDs, because these two effects lead to a different electric field and carrier density distribution across the device, affecting the position-dependent recombination probability. The present results show that for PPV-based diodes at room temperature, the density contribution is very significant and cannot be neglected, as it has been so far in the description of the charge transport in these devices. In general, the dominance of either carrier density or electric field will be a complex function of temperature, applied voltage, device geometry, and amount of disorder in the polymer. A theoretical model has recently been developed to disentangle all these effects.

In conclusion, we have investigated the hole transport in NRS-PPV hole-only devices as function of sample thickness. The SCL current in hole-only diodes can be governed by both the dependence of the hole mobility on the electric field and the charge carrier density. The thickness dependence enables us to discriminate between the two contributions. The experimentally obtained enhanced thickness dependence demonstrates that, for these polymeric LEDs at room temperature, the deviation of the SCL current at high voltages from the Mott–Gurney law is predominantly due to the dependence of the mobility on the carrier density.

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