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The electric field at hole injecting metal/organic interfaces as a cause for manifestation of exponential bias-dependent mobility

B. Cvikl *

Faculty of Energy Technology and Faculty of Civil Engineering, University of Maribor, 2000 Maribor, Slovenia
"Jožef Stefan" Institute, 1000 Ljubljana, Slovenia

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ABSTRACT

It is shown that the well-known empirical exponential bias-dependent mobility is an approximation function of the relevant term emerging in the Mott–Gurney space charge limited current model when the *constant* non-zero electric field at the *hole* injecting metal/organic interface E_{int} is taken into account. The term in question is the product of the bias-independent (but organic layer thickness-dependent) effective mobility coefficient and the algebraic function, $f(\lambda)$, of the argument $\lambda = E_{int}/E_a$, where E_a is the externally applied electric field. On account of the non-zero interfacial field, E_{int}, the singularity of the spatial dependence of the hole current density, $p(x)$, is removed. The resulting *hole drift* current density, j, is tested as a function of E_a against a number of published room temperature hole current j–E_a data sets, all characterized by good ohmic contact at the hole injecting interface. It is shown that the calculated current density provides a very good fit to the measurements within a high range of E_a intervals. Low values of E_a, are investigated analytically under the assumption of hole drift-diffusion. The extremely large internal electric fields at the anode/organic junction indicate drift-diffusion to be an improbable process for the structures investigated. However, a description of hole transport throughout the whole interval of experimental E_a values may be obtained at low values of E_a by an extended Mark–Helfrich drift model with traps occupying the exponentially distributed energy levels, followed by the extended Mott–Gurney model description within the remaining part of the E_a interval. In both models the same (bias-independent) effective mobility coefficient is incorporated into the calculations. The results present evidence that within the framework of the extended Mott–Gurney expression the properly derived term should replace the empirical exponential bias-dependent mobility, making it redundant in the interpretation of j–V data.

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1. Introduction

The performance of an organic electronic device depends substantially upon the charge injection at the metal/organic interface, the concentration of charge traps and the mobility of the charge carrier within the organic bulk. The investigation of charge transport in disordered organic semiconductors is of high importance for the understanding of these complicated processes purely on a fundamental basis, and very much so for device applications. Currently no comprehensive and self-consistent theory exists that is able accurately to predict their transport properties, in spite of the fact that many different model descriptions exist in the literature [1–3].

The charge transport properties are usually determined by considering current density–voltage curves measured on unipolar single layer organic structures, where interpretation of the data is based on fitting them with analytic equations based on the assumption of drift charge transport, such as the Mott–Gurney law [4] or the Mark–Helfrich equation [5]. However, recent numerical simulations have shown [6–11] that the current density in unipolar single layer organic structures is a combination of the drift-diffusion transport of the charge carriers. In particular it has been shown that the inclusion of charged traps into the calculations strongly enhances the importance of the diffusion term of the current, particularly at low values of the external bias. This is precisely the region where very often the predictions of the Mott–Gurney analytical expression are found to considerably overshoot the experimental data. As pointed out recently [11] this feature may occur due to omission of the diffusion term and the effect of traps (or alternatively the effect of built-in voltage) in the calculated current–voltage curve. Similarly, it was argued that the Mark–Helfrich model, which includes the effect of traps exponentially distributed within the transport band, might, owing to neglect of diffusion, either underestimate or overestimate the real tail slope to the current–voltage curve. In fact drift-diffusion charge transport in the presence of traps appears to reproduce "Poole–Frenkel"-like mobility behavior [4], even when assumed to be electric field-independent [11].

Recently Wang et al. [12] investigated hole injection from different metal oxides into the hole transporting organic semiconductor molecule *N,N*-diphenyl-*N,N*-bis-1-naphthyl-1-1-biphenyl-4,4-doamine (α-NPD), and presented arguments that any current–voltage, j–V, data

* Corresponding author.
E-mail address: bruno.cvikl@ijs.si.

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analysis (for example extraction of the charge carrier mobility) may lead to erroneous conclusions [13]. The authors [13] pointed out that the space charge limited current (SCLC) characteristics are expected to occur for a metal/organic injection barrier of, say, less than 0.25 eV, which is achieved by the good ohmic contact between the charge injecting organics and the metal electrode. In the present work the SCLC denotes the phenomenon of the electric current through an insulator being limited by the enhanced space charge accumulation (in excess of that present in thermal equilibrium) at the charge-injecting interface. In the literature the SCLC region is most often understood as the *j–V* region where the current density follows the expression which, written in the general form used nowadays, reads:

$$j = \frac{9}{8}\varepsilon\varepsilon_0\ \mu_0 \frac{E_a^2}{L} e^{\beta\sqrt{E_a}} \tag{1}$$

Here the relative permittivity of the organic compound is ε, ε_0 is the vacuum permittivity, E_a is the externally applied electric field $(E_a = V_a/L)$, L is the thickness of the organic layer within the structure, μ_0 is the zero field-free charge carrier mobility and β is the so-called Poole–Frenkel factor [4]. The last two parameters μ_0 and β defining the exponential bias dependence of the charge carrier mobility are often deduced from the current density–voltage, *j–V* relation, or by admittance spectroscopy (AS) methods [14,15]. For $\beta \equiv 0$ and $\mu_0 =$ (field independent) constant the current density–voltage curve of Eq. (1) has a slope of 2 on a log-log scale. This is in accordance with the Mott–Gurney law describing the case of no charged traps (or all traps full) within the organic. Murgatroyd [4] extended the Mott–Gurney law to the case of (shallow) traps occupying a single energy level, incorporating the Frenkel effect in the derivation, and showing that the mobility μ_0 is to be understood as the product of the free carrier mobility and the factor θ, the ratio of the free to the total charge in the organic layer. Gill [16] later presented evidence that the bias-dependent mobility of Eq. (1) is in fact unrelated to the Poole–Frenkel effect. Consequently, it was the empirical expression for the bias-dependent mobility, i.e.

$$\mu = \mu_0\ \exp\left(\beta E_a^{1/2}\right), \tag{2}$$

proposed by Gill [16], which was introduced in Eq. (1) in order to account for the additional (over and above E_a^2) contribution of the applied bias to the current density, as required by experiments. The validity of the combination expressed by Eq. (1) has been experimentally verified many times for amorphous materials and conjugated polymers. Consequently, this empirical expression for the charge carrier mobility, Eq. (2), is considered in the literature as an independent quantity unrelated to various current density–voltage models. The stated expression is still lacking a credible explanation [1–3] based on physical processes leading to the experimentally verified exponential bias dependent behavior of the (hole or electron) mobility.

The important characteristic of the SCLC model, Eq. (1), is the assumption of vanishing electric field at the metal/organic junction, with the consequence that the charge density at the charge-injecting interface diverges. This difficulty has traditionally been circumvented on the grounds that due to diffusion of charge carriers the electric field at this interface is likely to vanish [17]. However, on the basis of the analytical solution of the one dimensional drift-diffusion equation, this understanding was questioned recently on the basis that the finite charge density at the injection contact occurs on account of the drift of charge carriers [18] if an appropriate modification of the SCLC model is made. This suggestion is additionally confirmed below.

Eq. (1) is generally used for the determination of the charge carrier mobility within a given organic layer by the current–voltage, *j–V*, method [19–22]. These results have been compared to those determined by the independent time-of-flight, TOF, method. It is commonly considered that the methods are fully equivalent on account of the agreement in magnitude and in bias dependence of the charge carrier mobility within a given organic characterized by a good ohmic injection contact.

In this work, particular attention is given to Eq. (1), as the general representation of two important SCLC models, i.e. the Mott–Gurney law with no traps or traps completely filled, or the model with shallow traps occupying a single energy level within the organic hole transport band [4]. As stated above, both models incorporate the exponential bias-dependent mobility as an independent empirical expression. The purpose of this work is to gain an insight into the reason why Eq. (1) occasionally very well reproduces the unipolar *j–V* data from single layer metal/organic/metal structures within a broad interval of external bias [19], but more often its application is limited to the highest range only [20]. In this respect, the intention is to investigate the effect of the postulated non-zero electric field at the hole-injecting interface on the *j–V* line shape that results from such a modification of the model. The predictions were tested on two different sets of good ohmic contact metal/organic single-carrier structures [19,20] that exhibit considerable differences in their hole current–voltage line shapes.

It is shown that the introduction of the non-zero electric field at the hole injecting metal/organic interface, assuming the drift transport of holes, modifies the Mott–Gurney model of the unipolar current density within the single layer organic. As a result of the postulated non-zero interfacial electric field, $E(L)$, the spatial dependence of the hole current density, $p(x)$, turns out to be both finite and continuous within the boundaries of the organic. Based upon the data of Ref. [19] it is explicitly shown that the empirical exponential bias-dependent mobility may be understood as an approximation function of the derived algebraic function, $f(\lambda)$, of the parameter $\lambda = E(L)/E_a$, where $E(L)$ is the (*constant*) electric field at the hole injecting interface. The effective mobility, μ_{eff}, is bias-independent, but an organic layer thickness-dependent quantity in accordance with the experimental findings [20]. It is found that the thickness dependence is inversely correlated with the magnitude of $E(L)$ at the interface. It is shown that in such a way the extended Mott–Gurney model directly reflects the empirical bias-dependent mobility Eq. (2) if (EL) is bias-independent. It therefore provides the relationship of the experimental parameters μ_0, and β, with μ_{eff} and the magnitude of the electric field at the hole injecting metal/organic junction $E(L)$.

It is shown that the derived expression provides an excellent fit to the *j–V* data of Ref. [20] within the high range of the E_a interval only. Consequently, the low E_a data were additionally investigated on the basis of analytical solutions of the drift-diffusion equation. Since this mode of hole transport predicts the internal electric field at the anode/organic junction to be of the order of 10^6 greater than the one externally applied, its occurrence is improbable.

It is found that within the low bias region the group of *j–V* measurements of Ref. [20] is well explained in terms of the Mark–Helfrich SCLC model with traps occupying exponentially distributed energy levels, when extended by a *constant* non-zero interfacial electric field [23]. At a certain higher bias and beyond it this charge drift model transcends into the extended Mott–Gurney model description of hole charge transport as presented in this work. The calculated curves distinguish the fact that throughout the whole range of the applied bias the same bias-independent mobility coefficient is used.

For completeness it should be stated that the recent modeling [24] of the current transition from ohmic to SCLC characteristics in organic devices, is independent of the above, able to describe various experimentally determined current–voltage features, also on account of the finite charge density at the charge injecting interface.

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