

Charge transport and electrical properties in the organic small-molecule material NPB

L. G. WANG*, Y. X. GAO, X. L. LIU, L. F. CHENG

School of Electrical Engineering and Automation, Henan Polytechnic University, Jiaozuo, 454000, People's Republic of China

A systematic study of the charge transport and electrical properties in the organic small-molecule material N,N'-bis(1-naphthyl)-N, N'-diphenyl-1,1'-biphenyl-4,4'-diamine (NPB) has been performed. We show that the thickness dependent current density versus voltage characteristics of NPB hole-only devices can be accurately described using our recently introduced improved mobility model based on both the Arrhenius temperature dependence and non-Arrhenius temperature dependence. For the material studied, we find the model parameters of the width of the density of states $\sigma = 0.14$ eV and the lattice constant $a = 1.6$ nm. Furthermore, we show that the boundary carrier density has an important effect on the current density versus voltage characteristics. Too large or too small values of the boundary carrier density lead to incorrect current density versus voltage characteristics. The numerically calculated carrier density is a decreasing function of the distance to the interface, and the numerically calculated electric field is an increasing function of the distance to the interface. Both the maximum of carrier density and the minimum of electric field appear near the interface.

(Received October 31, 2015; accepted June 09, 2016)

Keywords: Charge transport, Electrical properties, Organic small-molecule semiconductor

1. Introduction

The most efficient white organic light-emitting diodes (OLEDs) that are currently developed for lighting applications are multilayer devices based on small-molecule organic semiconductors [1-4]. The approach toward optimizing the efficiency and lifetime is based on the introduction of novel layer structure concepts followed by empirical optimization. The most promising device structures are selected on the basis of the results from current-voltage-luminance measurements. It is essential to understand how the current density and charge carrier distribution in each layer depend on the layer structure and thicknesses, and on the applied voltage. This will help considerably to build up a predictive OLED device model that allows one to rationally design OLEDs with increased performance.

For developing a predictive OLED device model it is crucial to understand how the charge carriers in these small-molecule organic semiconductors are transported, in particular how the mobility μ depends on the temperature T , the electric field E , and the charge carrier density p [5]. In past decades, the dependence of the mobility μ on temperature T and electric field E has been extensively investigated [6-11]. Monte Carlo simulations by Bäessler *et al.* for the Gaussian disorder model (GDM) showed a non-Arrhenius temperature dependence $\mu \propto \exp[-(T_0/T)^2]$ and a Poole-Frenkel behavior $\mu \propto \exp[\gamma\sqrt{E}]$ for the electric field dependence in a limited field range [6, 7]. Gartstein and

conwell pointed out that in order to obtain a Poole-Frenkel behavior in a broad range of field strengths, it is necessary to assume correlation between the site energies, leading to the correlated disorder model (CDM) [8]. However, recently, it was recognized that the importance of another parameter had been overlooked: the charge-carrier density p [12, 13]. From a numerical solution of the master equation for hopping transport in a disordered energy system with a Gaussian density of states, a complete description of the charge transport has been developed by Pasveer *et al.* that determines the dependence of the mobility μ on temperature T , carrier density p , and electric field E , which is known as the "extended Gaussian disorder model" (EGDM) [14]. It has been demonstrated that the EGDM is more applicable for disordered organic semiconductors than the commonly used model within which the mobility depends only on the electric field [15, 16]. Nevertheless, it is worth noting that their model, only having a non-Arrhenius temperature dependence $\ln(\mu) \propto 1/T^2$, can well describe the charge transport at low carrier densities, whereas at higher carrier densities, it becomes no longer satisfactory. Recently, we proposed an improved unified description of the temperature, carrier density and electric field dependence of the mobility based on both the Arrhenius temperature dependence $\ln(\mu) \propto 1/T$ and the non-Arrhenius temperature dependence $\ln(\mu) \propto 1/T^2$ [17]. It has been demonstrated that the improved model is more applicable for disordered organic materials than the EGDM, especially at high carrier density and high electric

field [18-20].

In the present work, we apply the improved model to the experimental current density versus voltage ($J-V$) characteristics of hole-only devices made from organic small-molecule material N,N'-bis(1-naphthyl)-N,N'-diphenyl-1,1'-biphenyl-4,4'-diamine (NPB). This material is widely used in OLEDs as a hole-injection layer, hole-transport layer, electron-blocking layer, blue-emitting layer, and as a host material in mixed emitting layers. We show that a consistent and precise description of $J(V)$ curves can be obtained by using the improved model. Furthermore, we calculate and analyze some important electrical properties for NPB by using the improved model, including the variation of current density versus voltage characteristics with the boundary carrier density, and the distribution of charge carrier density and electric field with the position.

2. Experiment

The devices used to study the charge transport and electrical properties in NPB have the following structure: Glass | ITO | NPB | AL. The 100 nm thick indium tin oxide (ITO) coated glass substrates were prepared and cleaned with acetone and isopropyl alcohol. Before depositing organic small-molecule material NPB layers by thermal evaporator, the surface of ITO was treated with UV ozone for 10 min. The 100 nm, 200 nm and 300 nm thick NPB layers were deposited by evaporation in a high-vacuum system with a substrate temperature of 300 K, followed by deposition of a 100 nm aluminum cathode layer. The thicknesses of the deposited layers were monitored during deposition using a calibrated resonance crystal. The $J-V$ characteristics of the hole-only devices were measured by a Keithley 4200 semiconductor parameter analyzer. All devices were encapsulated prior to the measurement. In this device geometry, electron injection from the AL cathode to the hole-transport layer is prohibited due to the large injection barrier. No light emission was observed from the devices up to the highest voltages applied in this study.

3. Results and discussion

Using the improved model of the temperature T , carrier density p and electric field E dependence of the mobility μ and the numerical calculation method as described in Ref. 17-19, we now perform a systematic study of the charge transport and electrical properties for organic small-molecule material NPB. The improved mobility model has three main parameters: the site spacing a (lattice constant or hopping parameter), the width of the Gaussian distribution of density of states σ , and a prefactor for the mobility μ_0 , which defines the mobility in the limit of zero density, zero electric field and infinite temperature. The solution of the coupled equations describing the space-charge limited current with the

improved model and the experimental $J-V$ measurements for NPB hole-only devices with various layer thicknesses at room temperature are shown in Fig. 1. It can be seen from the figure that the thickness dependent $J-V$ characteristics of NPB hole-only devices can be excellently described using a single set of parameters, $a = 1.6$ nm, $\sigma = 0.14$ eV, and $\mu_0 = 1000$ m²/V s. The parameters a , σ , and μ_0 are determined in such a way that an optimal overall fit is obtained. It is clear that our calculated results are in good agreement with experimental data. This suggests that the improved model is suitable to study the $J-V$ characteristics of NPB, and captures the physical essence of the hole transport in this organic material. As for the parameters, we have investigated the sensitivity of the quality of the fits in Fig. 1 to variation of the parameter values. The parameter μ_0 decreases with decreasing the value of a and σ , and increases with increasing the value of a and σ . These values of the parameters indicate that the lower value of σ can be mainly attributed to the omission of the p dependence and the lower value of a can be mainly attributed to the overestimation of the E dependence.

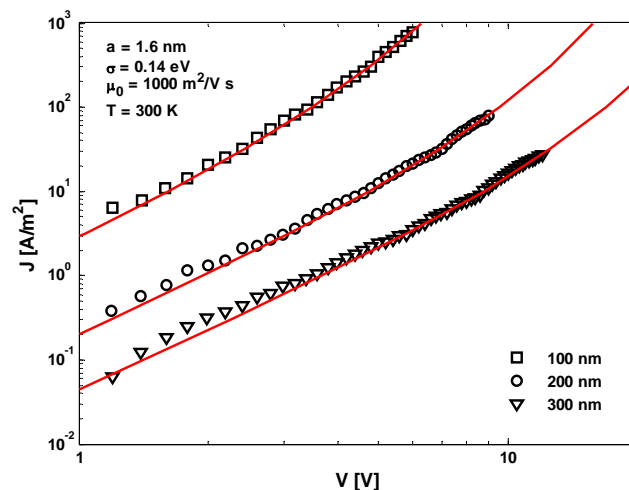


Fig. 1. Experimental results (symbols) and theoretical results (lines) of the current density versus voltage for $L=100, 200$ and 300 nm NPB layers at room temperature. The calculations are performed using our improved model, with parameters $a = 1.6$ nm, $\sigma = 0.14$ eV and $\mu_0 = 1000$ m²/V s.

It should be noted that the boundary carrier density (the carrier density at the interface) $p(0) = p_0$ is very important in the calculations, which may affect the $J-V$ characteristics. Thus, it is necessary to analyze the influence of the boundary carrier density $p(0)$ on the $J-V$ characteristics. The variation of $J-V$ characteristics with $p(0)$ for NPB hole-only devices with various layer thicknesses at room temperature are plotted in Fig. 2. The figure shows that the voltage is an increasing function of the current density, and the variation of voltage with $p(0)$ is dependent on the current density. The voltage decreases with increasing $p(0)$ for enough small value of $p(0)$ and shows a steep decrease with decreasing current density. On the other hand, the voltage increases with increasing $p(0)$ for sufficiently large values of $p(0)$ and shows a steep increase with

decreasing current density. Nevertheless, in the middle region, the $V - p(0)$ curves are fairly flat. This means that the voltage is almost independent of $p(0)$ in this region. Now we analyze the physical reason for variation in Fig. 2. In the small $p(0)$ region, the concentration of charge carriers in the bulk material is small, whereas the

impedance and voltage are relatively large. As the $p(0)$ increases into the middle region, the carrier injection near the interface and the drain into the bulk material will reach equilibrium, the $J - V$ characteristics will move into the Ohmic region and the $V - p(0)$ curves will become flat.

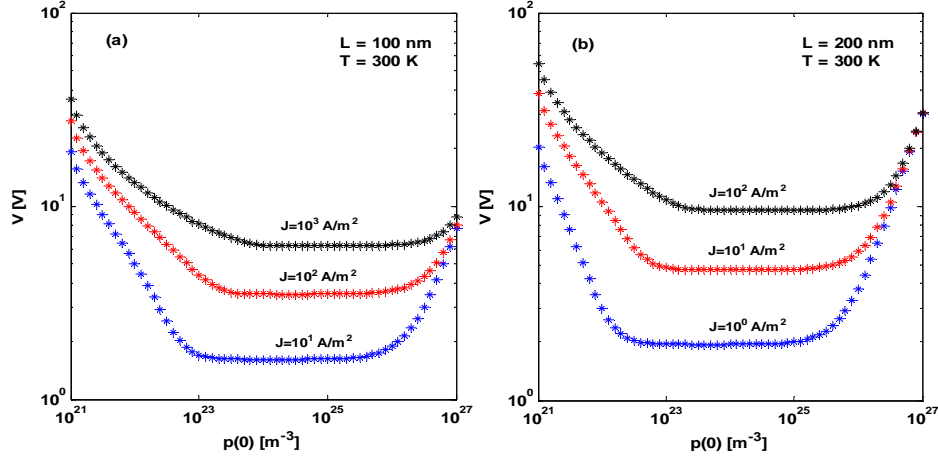


Fig. 2. Theoretical results of the voltage V versus the boundary carrier density $p(0)$ for a 100 nm and a 200 nm NPB layers at room temperature. Different Lines correspond to different current density values.

The calculated results of the carrier density as a function of position (the distance to the interface within the NPB layer) for NPB hole-only diodes are shown in Fig. 3. According to the above analysis of the influence of $p(0)$ on the $J - V$ characteristics, we take the values of $p(0)$, $(0.1, 0.5, 1) \times 10^{23} \text{ m}^{-3}$, for NPB in our calculations. The figure shows that the carrier density $p(x)$ is a decreasing function of the distance x . The speed of decrease is rapid for the cases with large values

of $p(0)$, whereas the speed of decrease is slow for small values of $p(0)$, and the corresponding curves are flat. As the distance x increases, $p(x)$ rapidly reaches the saturated values. Moreover, it can be seen from the figure that the thickness of accumulation layer (or decay distance) is a function of $p(0)$. The larger the value of $p(0)$, the smaller the thickness of accumulation layer, and vice versa.

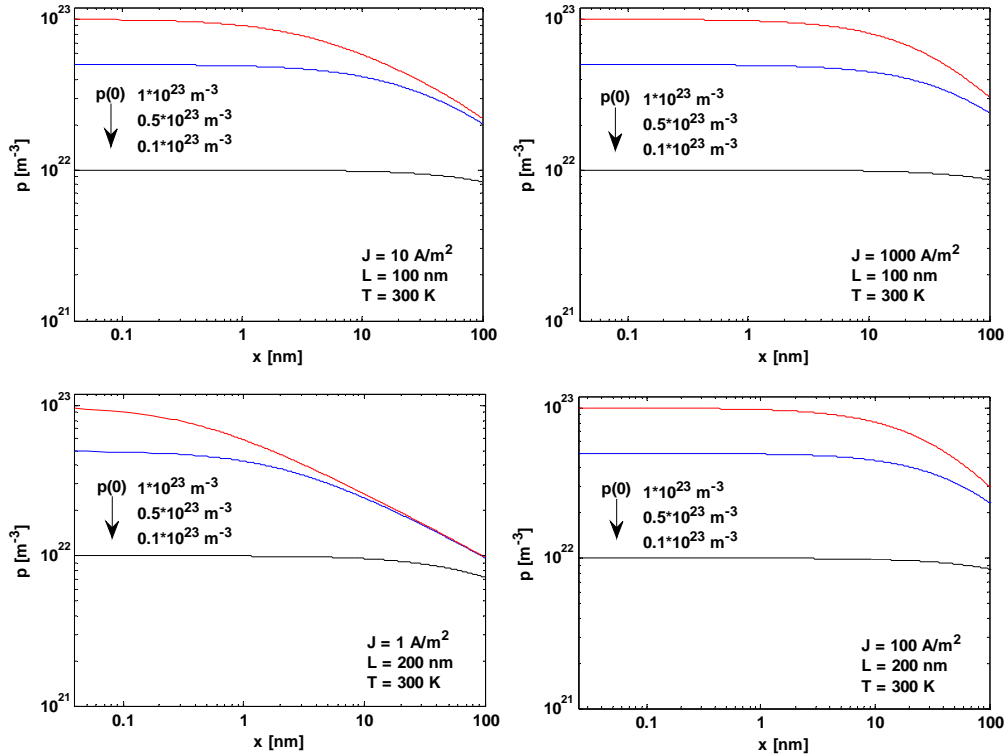


Fig. 3. Numerically calculated distribution of the charge carrier density p as a function of position x in NPB hole-only devices for low and high current densities at room temperature.

Fig. 4 shows the calculated results of the electric field as a function of position for NPB hole-only diodes. The figure shows that the electric field E is an increasing function of the distance x , and the shape of $E-x$ curves are similar to the reflection of $p-x$ curves in Fig. 3. The speed of increase is rapid for the cases with large values of $p(0)$. The thickness of accumulation layer deduced from the figure is consistent with Fig. 3.

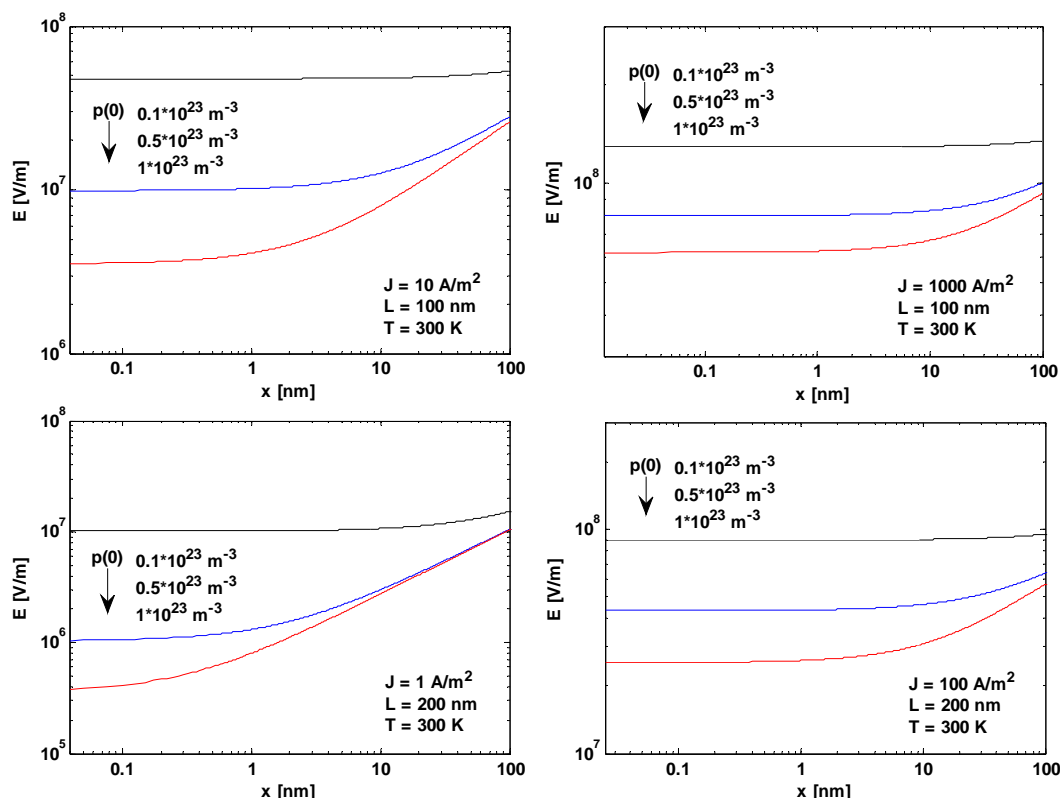


Fig. 4. Numerically calculated distribution of the electric field E as a function of position x in NPB hole-only devices for low and high current densities at room temperature.

4. Summary and conclusions

In summary, we have demonstrated that the $J-V$ characteristics of NPB hole-only devices can be accurately described using our improved mobility model based on both the Arrhenius temperature dependence and the non-Arrhenius temperature dependence. It is validated that our improved model is suitable for organic small-molecule material. For the material studied, the model parameters have realistic values, the width of the DOS $\sigma = 0.14$ eV and the lattice constant $a = 1.6$ nm. Furthermore, we show that the boundary carrier density has an important effect on the $J-V$ characteristics. Too large or too small values of the boundary carrier density lead to incorrect $J-V$ characteristics. The numerically calculated carrier density is a decreasing function of the distance to the interface, and the numerically calculated electric field is an increasing function of the distance to the interface. Both the maximum of carrier density and the minimum of electric field appear near the interface.

Conclusively, the results on $p(x)$ and $E(x)$ presented in Fig. 3 and Fig. 4 clearly exhibit the space-charge limited current density and voltage. The injection of carriers from electrode into NPB layer leads to carrier accumulation near the interface and decreasing function $p(x)$. The distribution $p(x)$ leads to the variation of $E(x)$, and the carrier accumulation near the interface results in increasing function $E(x)$.

Acknowledgements

This work is supported by the National Natural Science Foundation of China Grant No. 61501175, the Doctoral Scientific Research Foundation of Henan Polytechnic University Grant No. B2014-022 and Student's Platform for Innovation and Entrepreneurship Training Program Grant No. 201410460045.

References

- [1] B. W. D'Andrade, S. R. Forrest, *Adv. Mater.* **16**, 1585 (2004).
- [2] K. Walzer, B. Maennig, M. Pfeiffer, K. Leo, *Chem. Rev.* **107**, 1233 (2007).
- [3] J. K. F. So, P. Burrows, *MRS Bull.* **33**, 663 (2008).
- [4] S. Reineke, F. Lindner, G. Schwartz, N. Seidler, K. Walzer, B. Lüssem, K. Leo, *Nature (London)* **459**, 234 (2009).
- [5] S. L. M. van Mensfoort, V. Shabro, R. J. de Vries, R.

- A. J. Janssen, R. Coehoorn, *J. Appl. Phys.* **107**, 113710 (2010).
- [6] L. Pautmeier, R. Richert, H. Bässler, *Synth. Met.* **37**, 271 (1990).
- [7] H. Bässler, *Phys. Status Solidi B* **175**, 15 (1993).
- [8] Y. N. Gartstein, E. M. Conwell, *Chem. Phys. Lett.* **245**, 351 (1995).
- [9] D. H. Dunlap, P. E. Parris, V. M. Kenkre, *Phys. Rev. Lett.* **77**, 542 (1996).
- [10] S. V. Novikov, D. H. Dunlap, V. M. Kenkre, P. E. Parris, A. V. Vannikov, *Phys. Rev. Lett.* **81**, 4472 (1998).
- [11] Y. Preezant, N. Tessler, *Phys. Rev. B* **74**, 235202 (2006).
- [12] Y. Roichman, N. Tessler, *Synth. Met.* **135**, 443 (2003).
- [13] C. Tanase, E. J. Meijer, P. W. M. Blom, D. M. de Leeuw, *Phys. Rev. Lett.* **91**, 216601 (2003).
- [14] W. F. Pasveer, J. Cottaar, C. Tanase, R. Coehoorn, P. A. Bobbert, P. W. M. Blom, D. M. deLeeuw, M. A. J. Michels, *Phys. Rev. Lett.* **94**, 206601 (2005).
- [15] S. L. M. van Mensfoort, R. Coehoorn, *Phys. Rev. B* **78**, 085207 (2008).
- [16] S. L. M. van Mensfoort, S. I. E. Vulto, R. A. J. Janssen, R. Coehoorn, *Phys. Rev. B* **78**, 085208 (2008).
- [17] L. G. Wang, H. W. Zhang, X. L. Tang, C. H. Mu, *Eur. Phys. J. B* **74**, 1 (2010).
- [18] L. G. Wang, H. W. Zhang, X. L. Tang, Y. Q. Song, *Physica B* **406**, 68 (2011).
- [19] L. G. Wang, H. W. Zhang, X. L. Tang, Y. Q. Song, Z. Y. Zhong, Y. X. Li, *Phys. Scr.* **84**, 045701 (2011).
- [20] I. Katsouras, A. Najafi, K. Asadi, A. J. Kronemeijer, A. J. Oostra, L. J. A. Koster, D. M. de Leeuw, P. W. M. Blom, *Org. Electron.* **14**, 1591 (2013).

*Corresponding author: wangliguo@hpu.edu.cn