Effect of energetic disorder and diffusion on charge transport in organic solar cells

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In this paper, the effect of energy disorder and carrier diffusion on charge transport in the photovoltaic blend of small molecule donor BTR and fullerene acceptor $PC_{71}BM$ has been investigated. From an analysis of the $J-V$ characteristics of BTR:$PC_{71}BM$ devices by using the improved mobility model incorporating diffusion and the extended Gaussian disorder model (EGDM), it is observed that the improved model can better describe the charge transport properties and taking diffusion effect in photovoltaic devices into account is necessary. Furthermore, it is found that the carrier mobility in both hole-only and electron-only devices based on the BTR:$PC_{71}BM$ blend is closely related to energy disorder, and lower energy disorder corresponds to higher carrier mobility. This suggests that the amount of energetic disorder in organic solar cells appears to govern the charge transport properties.

(Received October 31, 2024; accepted June 3, 2025)

Keywords: Charge transport, Mobility model, Energy disorder, Diffusion, Organic solar cells

1. Introduction

With the rapid development of organic electronics, various highly promising organic electronic materials and devices have attracted significant attention. Organic photovoltaics, as a highly representative type of organic electronic device, has undergone numerous structural transformations. Among these, the introduction of bulk heterojunction structure has significantly improved the conversion efficiency of organic solar cells (OSCs) [1-6]. Small-molecule OSCs have been the focus of research due to their high molecular order, ease of purification, and excellent batch-to-batch reproducibility [7-9]. Based on regio-regular benzodithiophene-terthiophene-rhodanine (BTR) as the electron donor and [6,6]-phenyl C_{71}-butyric acid methyl ester ($PC_{71}BM$) as the electron acceptor, BTR:$PC_{71}BM$ with nearly perfect morphology of nanofiber network in the active layer has become the preferred material for small-molecule OSCs that can achieve optimal performance [10-12]. However, it is noteworthy that the conversion efficiency of small-molecule OSCs is still not ideal compared to polymer solar cells, which have recently achieved a breakthrough efficiency of 19% [13-16]. To further improve the efficiency of small-molecule OSCs, it is crucial not only to study the device structure from a macroscopic perspective but also, more importantly, to gain a deeper understanding of the properties of disordered organic semiconductor from a microscopic viewpoint, with particular emphasis on the issue of charge transport.

To comprehensively investigate the optoelectronic performance of BTR:$PC_{71}BM$ photovoltaic devices and achieve higher efficiency, it is essential to explore the charge transport properties of BTR:$PC_{71}BM$ blend. In bulk heterojunction organic photovoltaic devices, the effect of morphology in the active layer on charge transport has been widely studied [17-21]. It is found that solvent vapor annealing (SVA) can enhance the crystallinity of BTR:$PC_{71}BM$ blend and promote more distinct phase separation, thereby improving charge separation and transport within the device [22, 23]. On the other hand, Koster et al. conducted theoretical research on charge transport in organic materials and found significant differences in transport conditions between ordered and disordered materials [24]. However, they did not explicitly specify the relationship between mobility and electric field, temperature, and charge carrier density. Based on a numerically exact approach, a parametrization scheme for the corresponding mobility functional $\mu(T, p, F)$ was constructed by Pasveer et al., which is only based on the non-Arrhenius temperature dependence $\ln(\mu) \propto 1/T^2$ and is known as the extended Gaussian disorder model (EGDM) [25]. Although this model is widely used, it is also heavily criticized [26-30]. The model deviates significantly from the simulated results based on the master equation, particularly in the regions of high carrier densities and high electric fields. Therefore, the state of research related to the theoretical description of charge transport in disordered organic semiconductors can hardly be considered as satisfactory. Furthermore, when organic

semiconductors are situated between electrodes, the formation of current is not only due to the drift of charge carriers. Mobility is used to describe the ability of charge carriers to move under the influence of an electric field, and the diffusion of charge carriers will manifest under weak conditions. It is shown that the diffusion effect should also be considered in the charge transport process of organic semiconductors [31-34].

In this paper, to provide a better description of charge transport, we will propose an improved mobility model in which the diffusion effect is considered, and the temperature dependence of mobility is based on both the non-Arrhenius temperature dependence and Arrhenius temperature dependence $\ln(\mu) \propto 1/T$. The rest of the paper is organized as follows. In Section 2, we explain some relevant theories and methods, and determine an improved description of the dependence of mobility on the temperature, carrier density, and electric field. In Section 3, we will compare the theoretical results of current-voltage ($J-V$) characteristics obtained from our model and EGDM with experiments, respectively, and investigate the effect of energetic disorder and diffusion on charge transport in BTR:$PC_{71}BM$ blend. Finally, a summary and conclusions are given in Section 4.

2. Models and methods

For the description of charge transport in organic photovoltaic devices, the drift-diffusion models for electrons and holes are valid and rely typically on the simultaneous solution of the charge transport, continuity and Poisson equations. The basic equations used in this simulation are the Poisson equation, given by

$$\frac{\partial^2}{\partial x^2}\varphi(x) = \frac{q}{\varepsilon_0 \varepsilon_r}\left[n(x) - p(x)\right] \tag{1}$$

where q is the basic charge, φ is the electric potential, ε_0 is the vacuum dielectric constant, ε_r is the material's relative dielectric constant. n and p represent the carrier densities of electrons and holes, respectively, which are composed of mobile carriers and trapped charges. The drift-diffusion equations for electrons and holes are as follows:

$$J_n = -qn\mu_n \frac{\partial \varphi}{\partial x} + qD_n \frac{\partial}{\partial x} n \tag{2a}$$

$$J_p = -qn\mu_p \frac{\partial \varphi}{\partial x} - qD_p \frac{\partial}{\partial x} p \tag{2b}$$

The current density consists of a drift part caused by the electric field and diffusion current. The mobility μ depends on the electric field F, temperature T, and carrier density p. The diffusion coefficient D is related to the mobility and is given by the generalized Einstein relation, which is described further in the following section.

For hopping on a simple cubic lattice with uncorrelated Gaussian disorder, a full description of the mobility taking into account both the field and carrier density dependence was obtained by Pasveer et al. in the form of the EGDM [25]. In the EGDM, the mobility can be expressed as

$$\mu(T, p, F) = \mu(T, p) f(T, F) \tag{3}$$

with $\mu(T, p)$ and $f(T, F)$ in the form:

$$\mu(T, p) = \mu_0(T) \exp\left[\frac{1}{2}(\hat{\sigma}^2 - \hat{\sigma})(2pa^3)^{\delta}\right] \tag{4a}$$

$$\mu_0(T) = \mu_0 b_1 \exp(-b_2 \hat{\sigma}^2), \tag{4b}$$

$$\delta \equiv 2[\ln(\hat{\sigma}^2 - \hat{\sigma}) - \ln(\ln 4)]/\hat{\sigma}^2, \quad \mu_0 \equiv a^2 \nu_0 e/\sigma, \tag{4c}$$

$$f(T, F) = \exp\left\{0.44\left(\hat{\sigma}^{3/2} - 2.2\right)\left[\sqrt{1 + 0.8\left(\frac{eaF}{\sigma}\right)^2} - 1\right]\right\} \tag{5}$$

where $\mu(T, p)$ and $f(T, F)$ are density dependent and field dependent factor, respectively. The EGDM is sometimes considered universal, and is the basis for commercially available organic devices simulation software [35, 36]. However, the methodology followed to derive the above EGDM parametrization has been heavily criticized for giving an inadequate description of especially the field dependence of the mobility [26-30]. The model deviates significantly from the simulated results from the master equation, particularly in the regions of high densities and high electric fields.

To provide a better description of charge transport, we propose an improved mobility model in which the diffusion effect is considered, and the temperature dependence of mobility is based on both the Arrhenius and non-Arrhenius temperature dependence. It can be described as follows:

$$\mu(T, p) = \mu_0(T) \exp\left[\frac{1}{2}\left(\hat{\sigma}^2 - \hat{\sigma}\right)\left(2pa^3\right)^{\delta}\right] \tag{6a}$$

$$\mu_0(T) = \mu_0 m_0 \exp(m_1 \hat{\sigma} - m_2 \hat{\sigma}^2) \tag{6b}$$

$$\delta \equiv 2\hat{\sigma}^{-2}\left[\ln(\hat{\sigma}^2 - \hat{\sigma}) - \ln(1.39)\right], \mu_0 \equiv \frac{a^2 \nu_0 e}{\sigma} \tag{6c}$$

where $m_0 = 0.48 \times 10^{-9}$, $m_1 = 0.80$, $m_2 = 0.52$, and μ_0 is the mobility prefactor. $\hat{\sigma} \equiv \sigma / k_B T$ is the dimensionless

disorder parameter, k_B is the Boltzmann constant, σ is the width of the Gaussian density of states (DOS), a is intersite distance, e is the charge of the carriers, and ν_0 is the attempt frequency.

The dependence of the electric field F and the diffusion coefficient D are given by the following relation:

$$\mu(T,F,p)=\mu(T,p)^{g_1(T,F)}\exp\left[m_3 g_1(T,F)-m_3\right] \quad (7)$$

$$g_1(T,F)=\left[1+m_4(Fea/\sigma)^2\right]^{-1/2} \quad (8)$$

$$g_2(T,p)=\frac{p}{k_B T\dfrac{\partial p}{\partial E_f}} \quad (9)$$

$$D=\frac{k_B T}{q}\mu(T,F,p)g_2(T,p) \quad (10)$$

$$m_3 = c_0 + c_1 \ln(\hat{\sigma}^3 a) \quad (11a)$$

$$m_4 = 118.3 + 9.09\ln(\hat{\sigma}^3 a) \quad (11b)$$

$$c_0 = 40.54 - 109.5\hat{\sigma} + 210.35\hat{\sigma}^{-2} + 32.31\hat{\sigma}^{-3} \quad (12a)$$

$$c_1 = -0.23 - 0.12\hat{\sigma} + 0.045\hat{\sigma}^2 + 0.027\hat{\sigma}^3 \quad (12b)$$

where g_1 is the weak density-dependent function, g_2 is the dimensionless diffusion coefficient enhancement function, E_f is the Fermi level, m_3 and m_4 are the weak density-dependent parameters.

3. Results and discussion

In order to explore the effect of energy disorder and diffusion on charge transport, we investigate the temperature-dependent $J-V$ characteristics of hole-only and electron-only devices based on as-cast BTR:$PC_{71}BM$ blend. Fig. 1 and Fig. 2 show the $J-V$ characteristics of hole-only device with a thickness of 290 nm at different temperatures. The solid lines represent the numerical calculations from the improved model and EGDM, respectively. It is found that the temperature-dependent $J-V$ characteristics of BTR:$PC_{71}BM$ hole-only device can be well described using only one set of parameters: σ = 0.123 eV, μ_0 = 42 m^2/Vs, and a = 1.8 nm. Obviously, the improved model is in good agreement with the experimental results. Under low voltage, the current density calculated from the EGDM considering only drift is significantly lower than experiment measurements. Moreover, this phenomenon is not improved at different temperatures, and the difference decreases gradually with the increase of voltage. It can be seen that the improved model is more accurate for describing charge transport, and the influence of diffusion effect on charge transport in BTR:$PC_{71}BM$ hole-only device is significant and cannot be ignored.

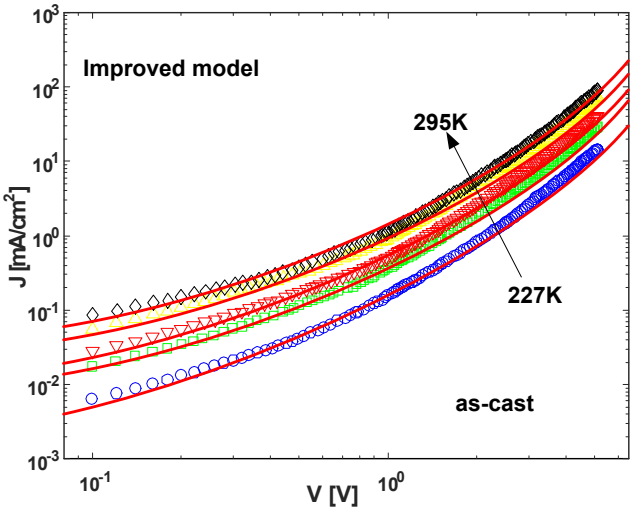

Fig. 1. Temperature dependent J-V characteristics of BTR:$PC_{71}BM$ hole-only device (as-cast). Symbols are experimental data from Ref. [11]. Lines are the numerical calculation results of the improved model (colour online)

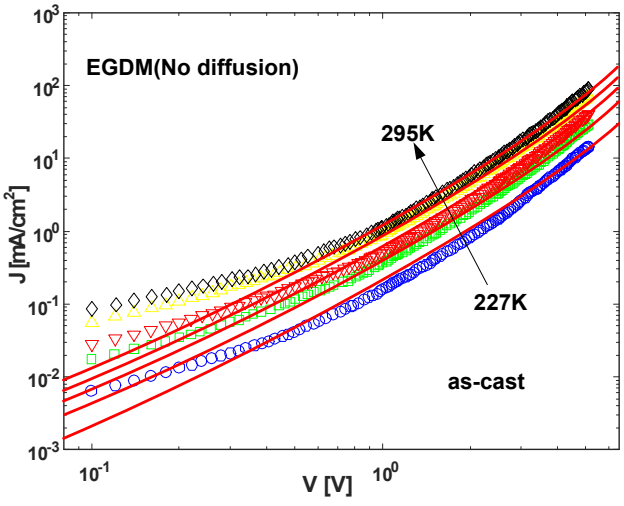

Fig. 2. Temperature dependent J-V characteristics of BTR:$PC_{71}BM$ hole-only device (as-cast). Symbols are experimental data from Ref. [11]. Lines are the numerical calculation results of the EGDM (colour online)

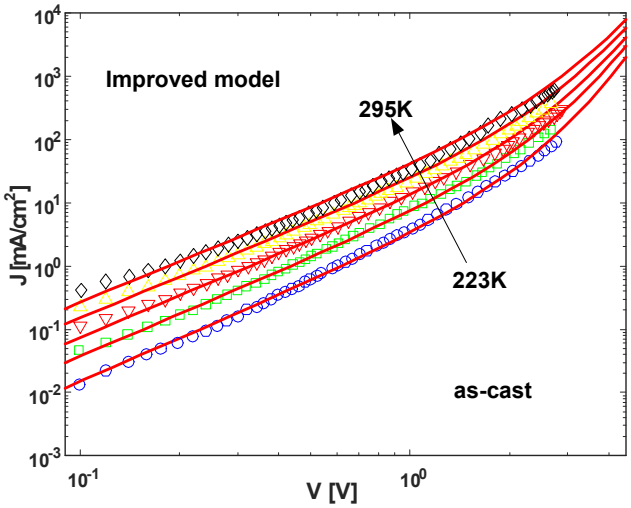

Fig. 3. Temperature dependent J-V characteristics of BTR:PC_{71}BM electron-only device (as-cast). Symbols are experimental data from Ref. [11]. Lines are the numerical calculation results of the improved model (colour online)

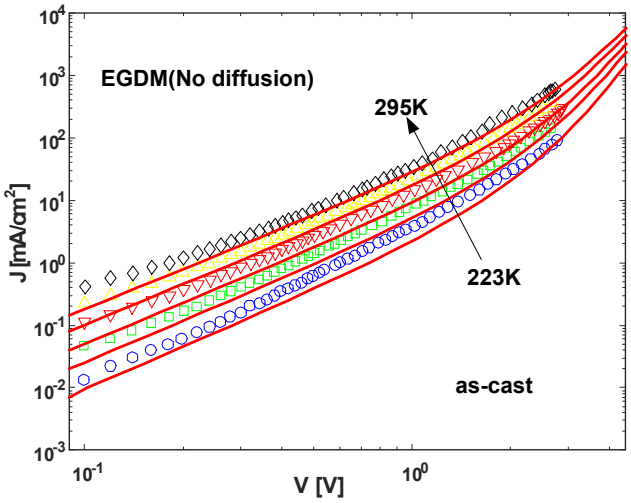

Fig. 4. Temperature dependent J-V characteristics of BTR:PC_{71}BM electron-only device (as-cast). Symbols are experimental data from Ref. [11]. Lines are the numerical calculation results of the EGDM (colour online)

Now we further consider whether diffusion also affects charge transport in BTR:PC_{71}BM electron-only device. When the above two models are applied to electron-only device, the temperature-dependent $J-V$ characteristics can also be well described with only one set of parameters: σ = 0.097 eV, μ_0 = 140 m^2/Vs, a = 1.75 nm. Fig. 3 and Fig. 4 clearly show that diffusion also has a significant impact on charge transport in electron-only device, with the improved model better describe charge transport than the EGDM. From Fig. 1 to Fig. 4, it is evident that the diffusion effect should be consider in the charge transport process of BTR:PC_{71}BM, and only considering drift cannot accurately describe the charge transport properties. Furthermore, it can be observed that the parameter σ values for BTR:PC_{71}BM hole-only and electron-only devices are 0.123 eV and 0.097 eV, respectively. The lower value of σ in the electron-only device indicates that the degree of energy disorder in electron transport is lower than in hole transport. The zero-field electron and hole mobility at room temperature of $6.9\times10^{-8}$$m^2$/Vs and $2.5\times10^{-9}$$m^2$/Vs can be obtained from the $J-V$ measurements (electron mobility is about 30 times higher than hole mobility). The imbalance of electron and hole mobility is detrimental to charge transport in BTR:PC_{71}BM blend, which will ultimately reduce device efficiency. This result aligns with the observation that the energy disorder for electron transport is lower than that for hole transport, indicating that a lower degree of energy disorder will result in higher mobility.

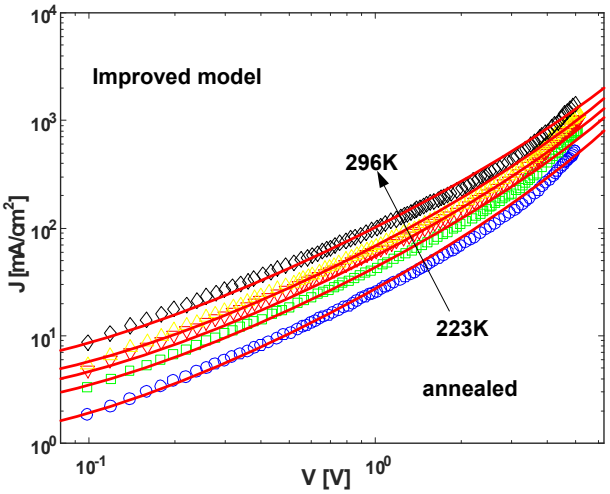

Fig. 5. Temperature dependent J-V characteristics of BTR:PC_{71}BM hole-only device (annealed). Symbols are experimental data from Ref. [11]. Lines are the numerical calculation results of the improved model (colour online)

Next, we will further investigate the effect of energy disorder and diffusion on charge transport in BTR:PC_{71}BM hole-only and electron-only devices under solvent vapor annealing (SVA). Fig. 5 to Fig. 8 display the temperature-dependent $J-V$ characteristics of hole-only and electron-only devices under solvent vapor annealing. The solid lines represent the numerical calculations from the improved model and EGDM. From Fig. 5 to Fig. 8, it can be seen that, for both hole-only and electron-only devices, the improved model fits the experimental data better than the EGDM. The parameters of hole-only and electron-only devices are σ = 0.088 eV, μ_0 = 270m^2/Vs, a = 1.48 nm and σ = 0.093 eV, μ_0 = 255 m^2/Vs, a = 1.2 nm, respectively. These results indicate that the improved model can also accurately describe the temperature-dependent $J-V$

characteristics of both hole-only and electron-only devices under annealing condition. Moreover, it reconfirms the necessity of considering diffusion effect, as relying solely on drift is inadequate. The zero-field electron and hole mobility under solvent vapor annealing are 1.25×10^{-7} m^2/Vs and 1.36×10^{-7} m^2/Vs, respectively. It can be seen that the mobility of electrons and holes under the annealing condition is greatly balanced, and the electron mobility is only slightly lower than that of holes, which also corresponds to the lower energy disorder for hole transport.

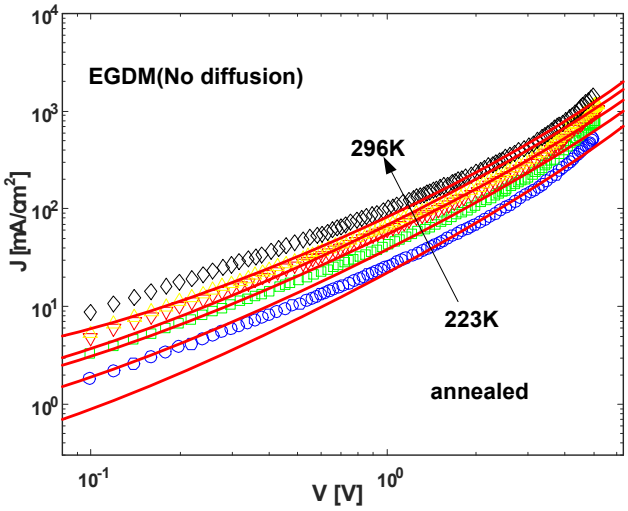

Fig. 6. Temperature dependent J-V characteristics of BTR:PC_{71}BM hole-only device (annealed). Symbols are experimental data from Ref. [11]. Lines are the numerical calculation results of the EGDM (colour online)

From Fig. 5 to Fig. 8, it can be found that under solvent vapor annealing conditions, both hole-only and electron-only devices based on BTR:PC_{71}BM exhibit higher mobilities compared to those under as-cast conditions, especially the hole mobility increases by nearly two orders of magnitude, bringing the mobilities of holes and electrons to almost equal levels. This can be interpreted as that solvent vapor annealing can improve the internal morphology of the material, promote charge extraction, increase crystallinity, and effectively improve the charge transport environment in BTR:PC_{71}BM blend [21, 22]. It is worth noting that the value of Gaussian energy disorder width for both hole-only and electron-only devices under solvent vapor annealing is smaller than that under as-cast conditions. For hole-only devices, σ decreases from 0.123 eV to 0.088 eV, while for electron-only devices, σ decreases from 0.097 eV to 0.093 eV. As the degree of energy disorder decreases, both hole and electron mobilities increase. These results indicate that lower energy disorder have higher carrier mobility, demonstrating a close relationship between carrier mobility and energy disorder. Using Gaussian disorder in the improved model can reduce the influence of many processes (such as trapping and capture effects) into a single disorder parameter. It is shown that the degree of energy disorder in disordered organic semiconductors appears to govern the charge transport properties.

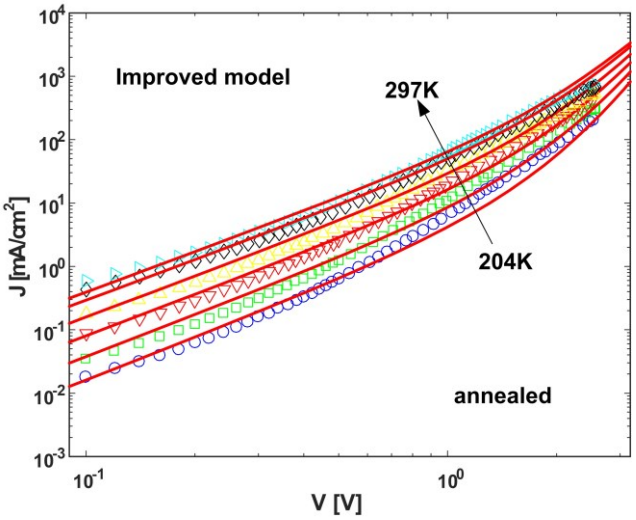

Fig. 7. Temperature dependent J-V characteristics of BTR:PC_{71}BM electron-only device (annealed). Symbols are experimental data from Ref. [11]. Lines are the numerical calculation results of the improved model (colour online)

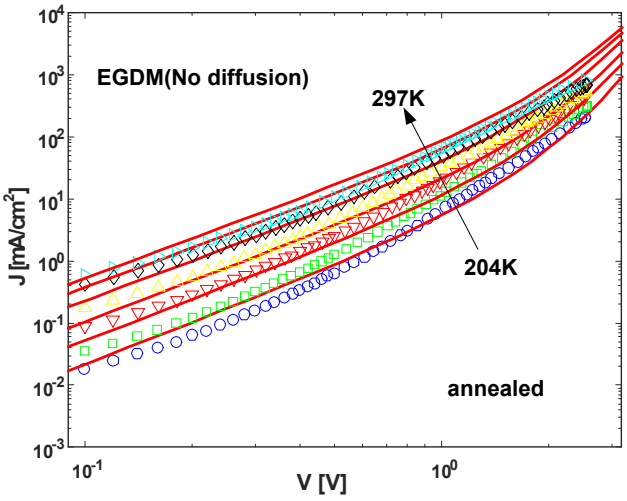

Fig. 8. Temperature dependent J-V characteristics of BTR:PC_{71}BM electron-only device (annealed). Symbols are experimental data from Ref. [11]. Lines are the numerical calculation results of the EGDM (colour online)

4. Summary and conclusions

In summary, charge transport in BTR:PC_{71}BM blend under as-cast and solvent vapor annealing is studied. It is found that the improved mobility model, which includes

diffusion effect, is more suitable for describing the charge transport characteristics in BTR:$PC_{71}BM$ blend. The diffusion effect in disordered organic semiconductors is significant and cannot be ignored. Furthermore, it is shown that lower energy disorder corresponds to higher hole and electron mobilities, indicating that energy disorder in disordered organic semiconductors appears to govern the charge transport properties. These results provide valuable information for exploring the effect of energy disorder and diffusion on the performance of organic photovoltaic devices.

Acknowledgements

This work is supported by the Natural Science Foundation of Henan Province Grant No. 242300420282 and No. 242300420685, the Science and Technology Project of Henan Province Grant No. 252102211107 and No. 242102241027, the Key Scientific Research Project of Colleges and Universities of Henan Grant No. 24A470006, the Fundamental Research Funds for the Universities of Henan Province Grant No. NSFRF240818 and No. NSFRF240712.

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