OPTOELECTRONICS AND ADVANCED MATERIALS – RAPID COMMUNICATIONS Vol. 18, No. 1-2, January-February 2024, p. 32 - 38

Effect of diffusion on charge transport in binary and ternary organic photovoltaic devices

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Recently, significant advances have been made in the field of organic solar cells (OSCs) containing high-performance active layer materials, electrodes, as well as novel device structures. Particularly, the use of ternary active layers and the innovation of non-fullerene acceptor materials have contributed significantly to the power conversion efficiency (PCE) improvement in OSCs. In this paper, we investigate the effect of diffusion on charge transport in binary and ternary organic photovoltaic blends. It is shown that the temperature dependent current density-voltage characteristics from the drift-diffusion simulations incorporating the extended Gaussian disorder model (EGDM) are more consistent with experimental data in comparison with those obtained from the only drift model in binary PBDB-T:IEICO-4F and ternary PBDB-T:IEICO-4F:$PC_{71}BM$ blends. Furthermore, it is found that the effect of diffusion on charge transport is more pronounced at low voltages and seems to be negligible when the applied voltage exceeds 1 V. The deviation of calculated curves from experimental measurements gradually increases with increasing temperature. It is of great importance to the influence of diffusion effect on charge transport in binary and ternary organic photovoltaic blends.

(Received August 22, 2023; accepted February 9, 2024)

Keywords: Charge transport, Organic photovoltaics, Diffusion, Ternary active layers

1. Introduction

In the last few decades, harnessing energy from the Sun is attracting increasing attention due to the challenges arising from our continued dependence on fossil fuels. Photovoltaic technologies are among the most efficient means of turning sunlight into direct current in solar cells. Among the various photovoltaic technologies, organic solar cells (OSCs) have drawn broad interest owing to their advantages such as being low cost, flexible, semitransparent, and ideal for roll-to-roll large-scale processing [1-5]. The first generation of OSCs was born with a single active layer, which was sandwiched between two electrodes with different work functions. However, the single layer devices showed poor power conversion efficiency (PCE) for the reason of difficulty in achieving efficient dissociation of excitons and severe recombination of electrons and holes. In 1986, a bilayer heterojunction structure was introduced by Tang [6], which was regarded as a big forward step in the field of OSCs. Nevertheless, the limited donor/acceptor (D/A) interface area still worked against the efficient exciton diffusion and separation, thus not yielding a high PCE in bilayer OSCs [7]. In 1995, the concept of the bulk-heterojunction (BHJ) architecture, where electron donating and electron accepting materials are blended together within an active layer, was introduced by Heeger et al. [8,9]. The BHJ structure presented an enhanced D/A interface and reduced the diffusion distance for exciton separation, resulting in a significant improvement in device performance. The invention of BHJ structure is a milestone in organic photovoltaic (OPV) research and addresses the critical issue present within the bilayer structure.

Although impressive efforts have been made in the last few decades (primarily in the binary BHJ architecture), due to intrinsic and extrinsic factors, efficiency and stability have not reached the benchmark for commercialization. Several significant endeavors have been done to improve efficiency and stability by means of modifying their active materials and device architecture [10-12]. One of the accomplishments to improve PCE while preserving flexibility, low cost and easy fabrication is the ternary system for BHJ active layer [13-17]. The binary system's narrow window of absorption spectra of organic materials reduces device efficiency, whereas the ternary system using multiple materials with different bandgaps improves light harvesting by broadening the spectral response of the active layer. A ternary system comprises a combination of a donor and two acceptors, or two donors and an acceptor, or a donor and an acceptor with a third component such as small molecules or dyes. Additional active components will make the BHJ system more complicated and challenging to understand; however, the benefit is very promising, which can efficiently enhance light absorption, optimize blend morphology, and facilitate charge or energy transfer through the additional component [18-20]. Because of the above mentioned superiorities, ternary OSCs have attracted a significant amount of attention and have experienced rapid progress with a PCE of over 18.0% [21-23]. Despite the progress in the PCEs, only limited research has been performed on charge transport in ternary OSCs. Charge transport directly impacts the performance of solar cells, while the electron and hole transport may also be relevant to exciton diffusion [24]. To date, the underlying physics and working mechanism of ternary OSCs is not yet fully understood and is still subject to debate.

In disordered organic semiconductors, charge motion is based on thermally activated hopping [25,26]. The most commonly used method to estimate the mobility of charge carriers and the underlying hopping and disorder parameters is that of space-charge limited currents (SCLC) [27-29]. In a single-carrier device with an Ohmic contact, the current will be limited by the transport in the bulk of the semiconductor, commonly known as an SCLC. In fact, when organic semiconductors are sandwiched between two electrodes, a metal-insulator-metal (MIM) diode stack is formed. The current that flows through such an organic semiconductor device, results from drift and diffusion of charge carriers. Drift and diffusion are related through mobility and diffusion coefficient. The mobility is used to describe the ability of carrier movement under the action of electric field, while diffusion coefficient is used to describe the ability of carrier movement in the presence of non-uniform concentration. The transport properties in the drift-dominated regime of such diodes have been extensively investigated, whereas the diffusion-dominated regime has rarely been taken into account so far. An important question is now whether the omission of diffusion effect is relevant in the analysis of charge transport in these organic semiconductors. In this paper, we investigate the effect of diffusion on charge transport in the ternary PBDB-T:IEICO-4F:PC$_{71}$BM (poly({4,8-bis[5-(2-ethylhexyl)-2-thienyl]benzo[1,2-b:4,5-b’]dithiophene-2,6-diyl}-2,5-thiophenediyl[5,7-bis(2-ethylhexyl)-4,8-dioxo-4H,8H-benzo[1,2-c:4,5-c’]dithiophene-1,3-diyl]):2,2’-[(2Z,2’Z)-({[4,4,9,9-tetr-akis(4-hexylphenyl)-4,9-dihydrosindaceno[1,2-b:5,6-b’]dithiophene-2,7-diyl]bis{4-[(2-ethylhexyl)oxy]thiophene-5,2-diyl}}bis(methanylylidene))bis(5,6-difluoro-3-oxo-2,3-dihydro-1H-indene-2,1-diylidene)]dimalononitrile):[6,6]-Phenyl C$_{71}$butyric acid methyl ester) blend, along with the binary polymer:non-fullerene PBDB-T:IEICO-4F blend. Firstly, a brief description of the model is given, followed by an overview of the relevant equations. Subsequently, we perform a detailed analysis of the temperature dependent current density-voltage ($J-V$) characteristics of hole-only device based on the binary PBDB-T:IEICO-4F blend, and hole-only and electron-only devices based on the ternary PBDB-T:IEICO-4F:PC$_{71}$BM blend by using the drift-diffusion simulations incorporating the extended Gaussian disorder model (EGDM), and the only drift model incorporating the EGDM, respectively.

2. Models and methods

For the description of charge transport in OPV devices, the general semiconductor drift-diffusion equations for electrons and holes are valid. Drift-diffusion models rely typically on the simultaneous solution of the charge transport, continuity and Poisson equations, while contacts are accounted for as boundary conditions. Only one spatial dimension is considered in this study, since OPV devices have a planar structure with a very small thickness compared to the lateral dimensions. The basic equations used in this simulation are the Poisson equation, given by

$$\frac{\partial^2}{\partial x^2}\psi(x) = \frac{q}{\varepsilon_0\varepsilon_r}[n(x) - p(x)] \qquad (1)$$

where q is the elementary charge, ε_0 is the vacuum permittivity, ε_r is the material’s relative dielectric constant, the electrical potential ψ is related to the electron and hole densities n and p, respectively. The carrier densities n and p consist of mobile charge carriers and trapped charges. The drift-diffusion equations for electrons and holes are

$$J_n = -qn\mu_n \frac{\partial}{\partial x}\psi + qD_n \frac{\partial}{\partial x} n \qquad (2a)$$

$$J_p = -qp\mu_p \frac{\partial}{\partial x}\psi - qD_p \frac{\partial}{\partial x} p \qquad (2b)$$

The current density consists of a drift part caused by the electric field and a diffusion current. The mobility μ is dependent on the electric field *F*, temperature *T* and carrier concentration *c*. The diffusion coefficient *D* is assumed to be related to the mobility and given by the generalized Einstein relation as discussed further below.

In this paper, we focus on charge transport in devices containing an organic semiconductor with Gussian disorder. The energy levels at neighboring sites are assumed to be uncorrelated. The Gaussian density of states (DOS) is described by

$$N(E) = \frac{N_0}{\sqrt{2\pi\sigma^2}}\exp\left[-\left(\frac{E-E_0}{\sqrt{2}\sigma}\right)^2\right] \qquad (3)$$

Where σ stands for the disorder parameter, i.e. the width of the Gaussian DOS. The parameter N_0 stands for the site density. In small-molecule organic semiconductors and in polymers, N_0 may be associated with the number of molecules and with the number of conjugated segments per volume unit, respectively. And the parameter E_0 denotes the reference energy level.

The disorder in organic semiconductors also affects the mobility function. A well-established mobility function that includes the effects of temperature *T*, carrier concentration *c*, and electric field *F* on the mobility μ is the extended Gaussian disorder model (EGDM) [30]. Previously, the EGDM has been successfully applied to describe charge transport in disordered organic semiconductors [25, 26, 31-33]. In the EGDM the mobility can be expressed as a product of a density-dependent and field-dependent factor

$$\mu(T, c, F) = \mu_0(T)g_1(T, c)g_2(T, F) \tag{4}$$

where $\mu_0(T)$ is the temperature-dependent mobility in the limit of a zero carrier density and zero electric filed.

$$\mu_0(T) = \mu_0 c_1 \exp(-c_2\hat{\sigma}^2), \tag{5a}$$

with $c_1 = 1.8 \times 10^{-9}$, $c_2 = 0.42$, $\hat{\sigma} \equiv \sigma / k_B T$, σ is the width of the Gaussian density of states (DOS). And g_1 and g_2 are dimensionless carrier density and field-dependent mobility enhancement factors, which are nonlinear and strongly increase with larger values of the disorder parameter.

$$g_1(T, c) = \exp[\frac{1}{2}\left(\hat{\sigma}^2 - \hat{\sigma}\right)\left(2pa^3\right)^{\delta}], \tag{5b}$$

$$g_2(T, F) = \exp\left\{0.44\left(\hat{\sigma}^{3/2} - 2.2\right)\left[\sqrt{1 + 0.8\left(\frac{eaF}{\sigma}\right)^2} - 1\right]\right\}, \tag{5c}$$

$$\delta \equiv 2\frac{\ln(\hat{\sigma}^2 - \hat{\sigma}) - \ln(\ln 4)}{\hat{\sigma}^2}, \quad \mu_0 \equiv \frac{a^2 \nu_0 e}{\sigma}, \tag{5d}$$

with a is the lattice constant, e is the charge of the carriers, and ν_0 is an attempt frequency.

In the EGDM, the Gaussian DOS also influences charge diffusion. Tessler et al. pointed out that the use of the generalized Einstein relation instead of the classical Einstein relation is correct [34, 35]. In this case the generalized Einstein diffusion coefficient is determined by

$$D = \frac{k_B T}{q}\mu(T, c, F)g_3(T, c) \tag{6}$$

where k_B is the Boltzmann constant and g_3 is a dimensionless diffusion coefficient enhancement function that follows from the shape of the density of states

$$g_3(T, c) = \frac{1}{k_B T}\frac{c}{\frac{\partial c}{\partial E_f}} \tag{7}$$

where E_f is the Fermi energy.

3. Results and discussion

In order to study the effect of diffusion on charge transport in binary and ternary organic photovoltaic systems, we investigate the temperature dependent current density-voltage ($J - V$) characteristics of single carrier devices based on binary PBDB-T:IEICO-4F and ternary PBDB-T:IEICO-4F:PC$_{71}$BM blends. To characterize charge transport, numerical drift-diffusion and only drift simulations are fitted to the experimentally temperature dependent $J - V$ characteristics. As we have confirmed in previous studies that the site energy in these organic photovoltaic blends is not correlated with the spatial location [36], we adopt the EGDM model with an uncorrelated Gaussian distribution of the random energies of hopping sites for simulation calculation. This model describes the mobility in the situation of hopping transport in a system with a Gaussian DOS distribution and includes the density dependence of the mobility as opposed to original GDM. The EGDM is a mobility function that described the temperature, field, and density dependence of the mobility based on three input parameters, viz. the width of the DOS distribution σ, the lattice constant a, and a mobility prefactor μ_0. By fitting the EGDM to charge transport measurements, these parameters can be reliably determined. Here, σ mainly controls the temperature and charge concentration dependence of the mobility, whereas a predominantly affects the field dependence of the mobility and μ_0 only influences the magnitude of the mobility.

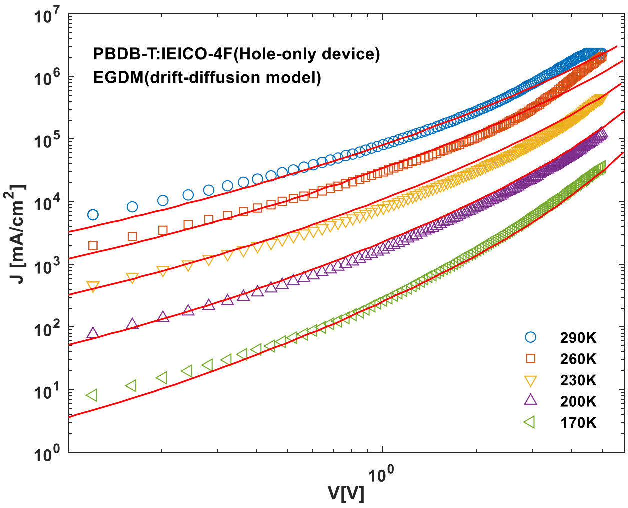

Fig. 1. Temperature dependent J-V characteristics of PBDB-T:IEICO-4F hole-only device. Symbols are experimental data from Ref. [37]. Lines are the numerically calculated results from the drift-diffusion model incorporating the EGDM (color online)

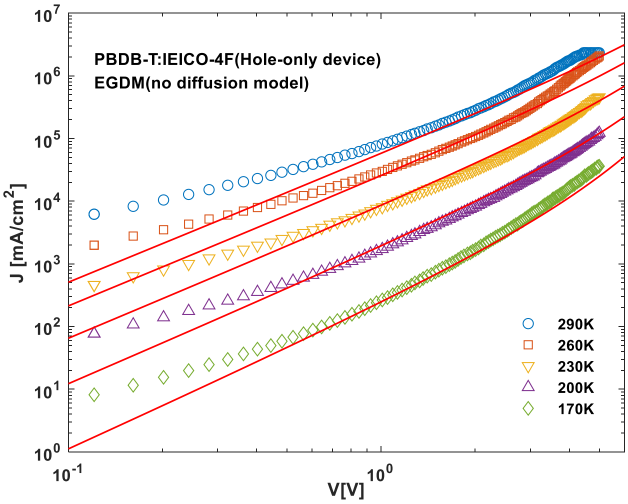

Fig. 2. Temperature dependent J-V characteristics of PBDB-T:IEICO-4F hole-only device. Symbols are experimental data from Ref. [37]. Lines are the numerically calculated results from the only drift model incorporating the EGDM (color online)

In view of the recent surge in non-fullerene acceptors, and the concomitant rise in PCEs in the organic photovoltaic field, we investigate the temperature dependent $J-V$ characteristics of the hole-only device based on polymer: non-fullerene PBDB-T:IEICO-4F blend by using both the drift-diffusion model incorporating the EGDM mobility function and the only drift model incorporating the EGDM. The solid lines in Fig. 1 and Fig. 2 represent the numerically calculated results from the drift-diffusion model incorporating the EGDM and the only drift model incorporating the EGDM, respectively. The symbols in Fig. 1 and Fig. 2 are the experimental measurements from Ref. [37]. It can be seen from Fig. 1 and Fig. 2 that the temperature dependent $J-V$ characteristics of PBDB-T:IEICO-4F hole-only device can be well described by using a single set of parameters, σ = 0.075 eV, a = 2.2 nm and μ_0 = 1200000 m^2/Vs. The parameters of σ, a, and μ_0 are determined in such a way that an optimal overall fit is obtained. What's more, it can be clearly found that the simulation curves calculated using the only drift model are not consistent with the experimental measurements at low voltages, the current density obtained is lower than the experimental measurements. The results present at various temperatures are very different, in which the deviation of calculated curves from experimental data gradually increases with increasing temperature. And it is not difficult to find that the calculated current density is always lower than the experimental measurements. However, as the voltage increases, this deviation becomes so small that it is almost negligible at high voltages (>1 V). On the other hand, as shown in Fig. 1, the fit quality of the drift-diffusion model to the experimental data is nearly perfect. From the above results, it can be concluded that the influence of diffusion effect on charge transport in PBDB-T:IEICO-4F hole-only device is significant and cannot be ignored.

Although the underlying working mechanisms are incompletely understood, ternary blend of one donor and two acceptors are proven to be a viable route to PCEs that exceed those of the corresponding binary compounds [13, 38]. It is therefore of interest to test the above two models on the ternary blend consisting of one donor and two acceptors. The temperature dependent $J-V$ characteristics of PBDB-T:IEICO-4F:PC$_{71}$BM hole-only device by using the drift-diffusion model incorporating the EGDM mobility function and the only drift model incorporating the EGDM are displayed in Fig. 3 and Fig. 4, respectively. The solid lines in Fig. 3 and Fig. 4 represent the numerically calculated results from the drift-diffusion model incorporating the EGDM and the only drift model incorporating the EGDM. The symbols in Fig. 3 and Fig. 4 are the experimental measurements from Ref. [37]. It can be seen from Fig. 3 and Fig. 4 that the temperature dependent $J-V$ characteristics of hole-only device based on PBDB-T:IEICO-4F:PC$_{71}$BM blend can be well described by using a single set of parameters, σ = 0.083 eV, a = 2.9 nm and μ_0 = 13000 m^2/Vs. By comparing the fit quality in Fig. 3 with Fig. 4, we can find that the numerically calculated results from the drift-diffusion model are more consistent with experimental data than the only drift model. The deviation of calculated curves from experimental measurements gradually increases with increasing temperature. The $J-V$ curves calculated from the only drift model at low voltage are obviously lower than the experimental measurements. However, as the voltage increases, this deviation becomes also so small that it is almost negligible at high voltages (>1 V). Therefore, it is not difficult to find in Fig. 3 and Fig. 4 that the influence of diffusion on the $J-V$ characteristics of PBDB-T:IEICO-4F:PC$_{71}$BM hole-only device is also notable.

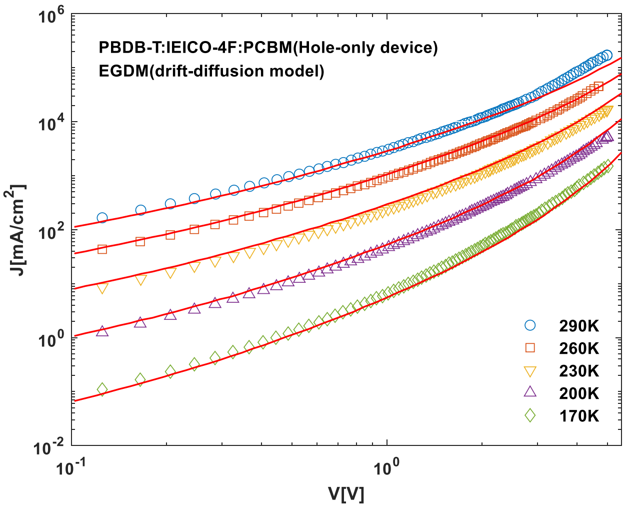

Fig. 3. Temperature dependent J-V characteristics of PBDB-T:IEICO-4F:PCBM hole-only device. Symbols are experimental data from Ref. [37]. Lines are the numerically calculated results from the drift-diffusion model incorporating the EGDM (color online)

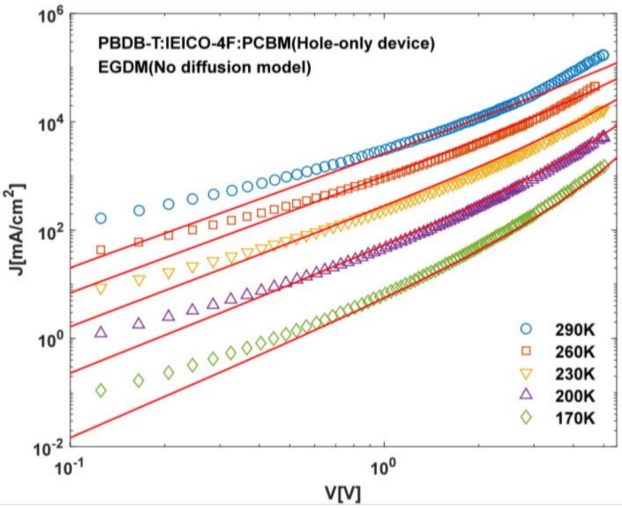

Fig. 4. Temperature dependent J-V characteristics of PBDB-T:IEICO-4F:PCBM hole-only device. Symbols are experimental data from Ref. [37]. Lines are the numerically calculated results from the only drift model incorporating the EGDM (color online)

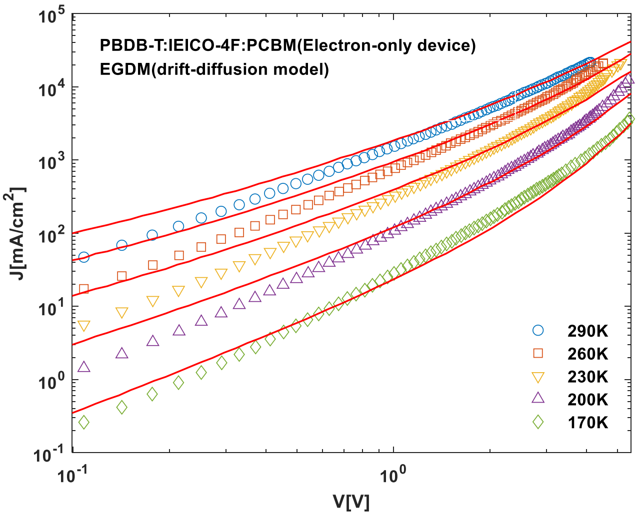

Fig. 5. Temperature dependent J-V characteristics of PBDB-T:IEICO-4F:PCBM electron-only device. Symbols are experimental data from Ref. [37]. Lines are the numerically calculated results from the drift-diffusion model incorporating the EGDM (color online)

We now consider the question whether the diffusion also affects charge transport in electron-only device based on PBDB-T:IEICO-4F:PC$_{71}$BM blend. When the above two models are also applied to electron-only device, it is not difficult to find in Fig. 5 and Fig. 6 that the influence of diffusion on the $J - V$ characteristics is also notable. It is clear from Fig. 5 and Fig. 6 that the temperature dependent $J - V$ characteristics of electron-only device based on PBDB-T:IEICO-4F:PC$_{71}$BM blend can also be well described by using a single set of parameters, σ = 0.06 eV, a = 3.6 nm and μ_0 = 950 m^2/Vs. The simulation curves calculated using the only drift model in Fig. 6 is lower than the experimental measurements at low voltages (< 1 V), whereas the numerically results from the drift-diffusion model in Fig. 5 is higher than the experimental data at low voltages (<1 V). On the other hand, the results present at various temperatures are very different, in which the deviation of calculated curves from experimental data gradually increases with increasing temperature for both drift-diffusion and only drift models.

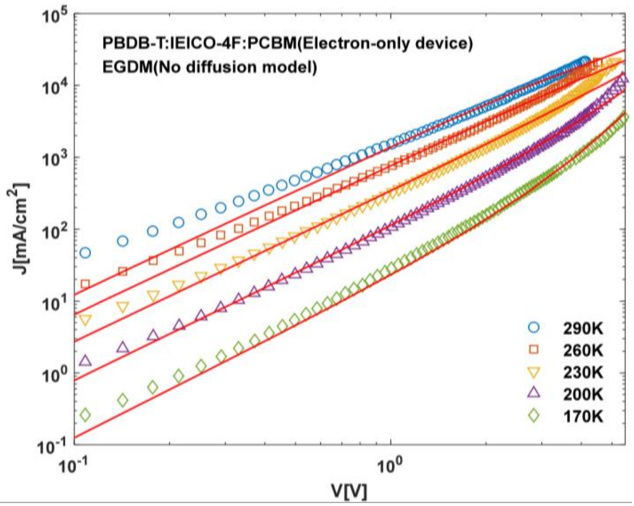

Fig. 6. Temperature dependent J-V characteristics of PBDB-T:IEICO-4F:PCBM electron-only device. Symbols are experimental data from Ref. [37]. Lines are the numerically calculated results from the only drift model incorporating the EGDM (color online)

It can be seen from Figs. 3-6 that the fit quality of EGDM to the $J - V$ characteristics of hole-only device based on PBDB-T:IEICO-4F:PC$_{71}$BM blend is better than electron-only device. A factor that may hamper the accurate description for the $J - V$ characteristics is various types of charge carrier traps, especially for electrons. We note that the disorder parameters of this ternary system are rather unremarkable in the range of 50-90 meV, with σ_{LUMO} typically being somewhat smaller than that of σ_{HOMO} and substantially smaller than what is typically found for PCBM [39, 40]. In addition, We find that the addition of PCBM to the binary system PBDB-T:IEICO-4F does not seem to lead to an appreciable increase in σ. On the other hand, we can find that the numerically results from the drift-diffusion model are more consistent with experimental data than the only drift model. It is worth noting that for single carrier devices based on binary PBDB-T:IEICO-4F and ternary PBDB-T:IEICO-4F:PC$_{71}$BM blends, ignoring the diffusion effect will have a significant impact on charge transport, which is reflected in the $J - V$ characteristics, especially at very low bias (<1 V). At higher voltages (>1 V), the effect of diffusion on charge transport can be disregarded.

4. Summary and conclusions

In summary, the effect of diffusion on charge transport in the binary PBDB-T:IEICO-4F blend and the ternary PBDB-T:IEICO-4F:PC_{71}BM blend is investigated. The temperature dependent $J-V$ characteristics of single carrier devices based on PBDB-T:IEICO-4F and PBDB-T:IEICO-4F:PC_{71}BM blends can be well described using the EGDM, the numerically results from the drift-diffusion model are more consistent with experimental data in comparison with those obtained from the only drift model. It is worth noting that ignoring the diffusion effect will have a significant impact on charge transport, which is reflected in the $J-V$ characteristics, especially at very low bias (＜1 V). This work not only proves the feasibility of describing charge transport in binary and ternary organic photovoltaic systems by using the EGDM, but also provides evidence on the need to take the effect of diffusion on charge transport into account in the PBDB-T:IEICO-4F and PBDB-T:IEICO-4F:PC_{71}BM blends.

Acknowledgements

This work is supported by the Fundamental Research Funds for the Universities of Henan Province Grant Nos. NSFRF200304 and NSFRF210424, the Young Key Teacher Program of Henan Polytechnic University Grant No. 2019XQG-17, the Key Scientific Research Project of Colleges and Universities of Henan Grant No. 24A470006, and the Doctoral Scientific Research Foundation of Henan Polytechnic University Grant Nos. B2014-022 and B2017-20.

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