

# The effect of spatially correlated disorder on the hole transport in PSF-TAD copolymers

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In this paper, the hole transport and spatial correlation between the site energies in blue light-emitting polyspirobifluorene (PSF) with copolymerized  $N, N, N', N'$  tetraaryldiamino biphenyl (TAD) hole transport units are investigated. It is shown that the temperature dependent and TAD concentration dependent current density versus voltage characteristics of PSF hole-only devices can be accurately described by using the improved extended Gaussian disorder model (IEGDM) and the extended correlated disorder model (ECDM), within which the mobility depends on the electric field and carrier density and within which spatial correlations between the site energies are absent or are included, respectively. Based on a comparison of the model parameters as obtained from both models, we view the more realistic intersite distance obtained using the IEGDM (1.3nm) compared to the value obtained using the ECDM (0.24nm) as an indication that in the PSF-TAD copolymers studied correlations between the site energies are absent or play a minor role.

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## 1. Introduction

Since their discovery, semiconducting conjugated polymers have been investigated extensively because of potential advantages like easy and cheap processing methods for fabrication of electronic devices [1–4]. Polymer light-emitting diodes (PLEDs) nowadays have started to emerge in display applications and lighting applications. Efficient blue emitters are essential for the realization of these applications. Polyfluorenes (PFs) are attractive blue emitters due to their wide band gap and high efficiency [5, 6]. Functionalizing these polymers in different structures has led researchers to optimize their stability and efficiency. Polyspirobifluorene (PSF) is a polymer from the PFs family in which the spiro center links two fluorene units to each other, making the polymer more stable and more efficient for light emission [7, 8]. A critical issue for the performance of PLEDs is the charge balance. However, polyfluorene derivatives often have a high ionization potential resulting in a large energy barrier for hole injection [9]. To improve the injection and conduction of holes, polyfluorene derivatives are often copolymerized with hole transport units (called guests) like arylamines [10, 11]. Furthermore, the hole transport in these copolymers can be tuned by varying the arylamine content, under the condition that their highest occupied molecular orbital (HOMO) level is higher than that of the host polymer [12]. At low concentrations the amine units act as hole traps and reduce the hole current, whereas above a critical concentration (typically 3–10%), the hole

transport from guest to guest becomes possible, leading to an increase of the mobility with increasing amine concentration [13–15]. As a result, the device operation of such a copolymer-based PLED is more complicated than for the conventional PLEDs. A detailed understanding of the charge transport in these copolymers is a prerequisite for the improvement of device performance.

The most important parameter characterizing the charge transport in disordered semiconducting polymers is the charge carrier mobility  $\mu$ . In the early modeling introduced by Bässler et al. [16, 17], the random energies were described by a Gaussian density of states (DOS), leading to the Gaussian disorder model (GDM). In this model, it is assumed that there is no spatial correlation between the site energies. Alternatively, it was suggested that the presence of dipole moments can give rise to spatial correlation between the site energies [18, 19], leading to the correlated disorder model (CDM). Later, it was realized that, apart from the dependence of the mobility  $\mu$  on the electric field  $E$  and temperature  $T$ , there is a strong dependence on the carrier density  $p$  [20–22], giving rise to extended versions of the GDM and CDM, the EGDM [23] and ECDM [24], respectively. Within the EGDM, the carrier density dependence of the mobility is much stronger than in the ECDM, but the electric field dependence is somewhat smaller. Furthermore, it should be noted that the EGDM, having a non-Arrhenius temperature dependence  $\ln(\mu) \propto 1/T^2$ , can only well describe the charge transport at low carrier densities. In order to better describe the charge transport, we proposed

an improved model in which the mobility depends on the temperature, electric field, and carrier density based on both the non-Arrhenius temperature dependence and Arrhenius temperature dependence  $\ln(\mu) \propto 1/T$  [25], which is known as the improved extended Gaussian disorder model (IEGDM). It has been demonstrated that the improved mobility model can rather well describe the charge transport in various organic materials [26-28]. Being able to make a distinction between various models, to determine the type of disorder and to accurately extract the materials parameters that determine the mobility in disordered organic semiconductors is of great importance to the rational design of organic light-emitting diodes (OLEDs). However, analyses of measured current density versus voltage ( $J - V$ ) curves of sandwich-type devices based on several polymers have been successfully carried out using the EGDM [29-32], whereas for several small-molecule materials, a more consistent analysis was obtained by using the ECDM [33, 34]. To date, it has remained unclear to what extent it is possible to make a distinction between the EGDM and ECDM, and the type of disorder (random or spatially correlated).

In this paper, the possible presence of spatially correlated disorder and the  $J - V$  characteristics of the hole transport for the blue-emitting PSF host and PSF with a varying number of  $N, N', N'', N'''$  tetraaryldiamino biphenyl (TAD) hole transport units host-guest copolymers are investigated. It is found that equally good descriptions of the temperature dependent and TAD concentration dependent  $J - V$  characteristics of hole-only devices based on PSF and PSF-TAD copolymers can be obtained within the IEGDM and ECDM, but a more realistic value of the intersite distance is obtained within the IEGDM than within the ECDM. This is an indication that in PSF and PSF-TAD copolymers spatially correlation between the site energies is absent or plays a minor role.

## 2. Models and methods

Based on the extended Gaussian disorder model (EGDM) [23], we recently proposed an improved mobility model (IEGDM) in which the dependence of the mobility  $\mu$  on the electric field  $E$ , carrier density  $p$ , and temperature  $T$  can be described as follows [25]:

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

$$\mu_0(T) = \mu_0 c_1 \exp(c_2 \hat{\sigma} - c_3 \hat{\sigma}^2), \quad (1b)$$

$$\delta \equiv 2 \frac{\ln(\hat{\sigma}^2 - \hat{\sigma}) - \ln(\ln 4)}{\hat{\sigma}^2}, \quad \mu_0 \equiv \frac{a^2 v_0 e}{\sigma}, \quad (1c)$$

with  $c_1 = 0.48 \times 10^{-9}$ ,  $c_2 = 0.80$ , and  $c_3 = 0.52$ . Where  $\mu_0(T)$  is the mobility in the limit of zero carrier density and electric field,  $\hat{\sigma} \equiv \sigma/k_B T$  is the dimensionless disorder parameter,  $\sigma$  is the width of the Gaussian

density of states (DOS),  $a$  is the lattice constant (intersite distance),  $e$  is the charge of the carriers, and  $v_0$  is the attempt frequency.

$$\mu(T, p, E) = \mu(T, p)^{g(T, E)} \exp[c_4(g(T, E) - 1)], \quad (2)$$

$$g(T, E) = [1 + c_5 (Eea/\sigma)^2]^{-1/2}, \quad (3)$$

where  $g(T, E)$  is a weak density dependent function,  $c_4$  and  $c_5$  are weak density dependent parameters, given by

$$c_4 = d_1 + d_2 \ln(pa^3) \quad (4a)$$

$$c_5 = 1.16 + 0.09 \ln(pa^3) \quad (4b)$$

$$d_1 = 28.7 - 36.3\hat{\sigma}^{-1} + 42.5\hat{\sigma}^{-2} \quad (5a)$$

$$d_2 = -0.38 + 0.19\hat{\sigma} + 0.03\hat{\sigma}^2 \quad (5b)$$

The extended correlated disorder model (ECDM) can be described as follows [24]:

$$\mu(T, p, E) = [(\mu_{low}(T, p, E))^{q(\hat{\sigma})} + (\mu_{high}(p, E))^{q(\hat{\sigma})}]^{1/q(\hat{\sigma})}, \quad (6)$$

$$q(\hat{\sigma}) = 2.4/(1 - \hat{\sigma}), \quad (7)$$

with  $\mu_{low}(T, p, E)$  the mobility in the low-field limit (the average reduced field  $E_{red} = eaE/\sigma \leq 1$ ), and with  $\mu_{high}(p, E)$  the mobility in the high-field limit (the average reduced field  $E_{red} = eaE/\sigma \geq 1$ ).

$$\mu_{low}(T, p, E) = \mu_0(T) g(T, p) f(T, E, p), \quad (8)$$

where  $g(T, p)$  and  $f(T, E, p)$  are the dimensionless mobility enhancement functions. These functions can be written as follows:

$$\mu_0(T) = 1.0 \times 10^{-9} \mu_0 \exp(-0.29\hat{\sigma}^2), \quad (9)$$

$$g(T, p) = \begin{cases} \exp[(0.25\hat{\sigma}^2 + 0.7\hat{\sigma})(2pa^3)^\delta], & pa^3 < 0.025 \\ g(T, 0.025a^{-3}), & pa^3 \geq 0.025 \end{cases}, \quad (10)$$

$$\delta \equiv 2.3 \frac{\ln(0.5\hat{\sigma}^2 + 1.4\hat{\sigma}) - 0.327}{\hat{\sigma}^2}, \quad (11)$$

$$f(T, E_{red}, p) = \exp[h(E_{red})(1.05 - 1.2(pa^3)^{r(\hat{\sigma})}) (\hat{\sigma}^{3/2} - 2)(\sqrt{1 + 2E_{red}} - 1)], \quad (12)$$

$$h(E_{red}) = 1, \quad r(\hat{\sigma}) = 0.7\hat{\sigma}^{-0.7}, \quad (13)$$

Within the very low-field,  $0 \leq E_{red} < 0.16 \equiv E_{red}^*$ ,

$h(E_{red})$  can be written as

$$h(E_{red}) = \begin{cases} \frac{4}{3} \frac{E_{red}}{E_{red}^*}, & (E_{red} \leq E_{red}^*/2) \\ \left[ 1 - \frac{4}{3} \left( \frac{E_{red}}{E_{red}^*} - 1 \right)^2 \right], & (E_{red}^*/2 \leq E_{red} \leq E_{red}^*) \end{cases}, \quad (14)$$

$$\mu_{high}(p, E) = \frac{2.06 \times 10^{-9}}{E_{red}} \mu_0 (1 - pa^3). \quad (15)$$

By using the above two mobility models, the  $J-V$  characteristics of organic electron devices can be exactly calculated by numerically solving the following equations adopting a particular uneven discretization method introduced in our previous paper [35, 36].

$$J = p(x)e\mu(T, p(x), E(x))E(x), \quad (16a)$$

$$\frac{dE}{dx} = \frac{e}{\epsilon_0 \epsilon_r} p(x), \quad (16b)$$

$$V = \int_0^L E(x) dx, \quad (16c)$$

where  $x$  is the distance from the injecting electrode,  $L$  is the organic semiconductor layer thickness sandwiched between two electrodes,  $\epsilon_0 \epsilon_r$  is the permeability of the organic materials.

### 3. Results and discussion

To study the hole transport and spatial correlation between the site energies for the PSF-TAD host-guest systems in a systematic way, the investigation of the temperature dependent and TAD concentration dependent  $J-V$  characteristics of PSF hole-only devices is performed by using the IEGDM and ECDM, respectively. Figure 1 shows the hole transport for a PSF hole-only device with 300 nm thickness at different temperatures. The hole transport is modeled using the IEGDM and numerical calculation method as described in Sec. 2. For the PSF polymers with 0% TAD, a single set of parameters  $a = 1.3$  nm,  $\sigma = 0.125$  eV, and  $\mu_0 = 250$  m<sup>2</sup>/Vs can consistently describe the hole transport. In an earlier study for the PSF-TAD copolymers, Nicolai et al. demonstrated that the hole transport in the copolymers with TAD concentrations ranging from 5–12.5 molar percent is trap-free space charge limited transport, and can be described by the extended Gaussian disorder model (EGDM) [23]. In order to better study the hole transport in PSF-TAD copolymers, the IEGDM is used to describe the hole transport in the PSF polymers with 0%, 5%, 7.5%, 10% and 12.5% TAD concentration. Figure 2 shows the  $J-V$  characteristics of all hole-only

devices for 0%, 5%, 7.5%, 10% and 12.5% TAD copolymers with thickness around 200 nm. As is clear from the figure for the 5% TAD copolymer, the mobility drops an order of magnitude as compared to the 0% TAD. The disorder parameter  $\sigma$  is 0.125 eV and the intersite distance is 1.3 nm for all copolymers, same as the parameters obtained in Fig. 1. In addition, it is worth noting that the value of the disorder parameter  $\sigma$  and the intersite distance  $a$  are significantly smaller than what were obtained by using the EGDM [14]. As for the parameter  $\mu_0$ , it ranges from 250, 35, 65, 75 and 165 m<sup>2</sup>/Vs for 0%, 5%, 7.5%, 10% and 12.5 % TAD concentration, respectively. Apparently, it can be observed from Fig. 1 and Fig. 2 that our calculated results are in good agreement with the experimental  $J-V$  measurements. This suggests that the IEGDM is suitable to study the charge transport in the PSF-TAD host-guest systems.

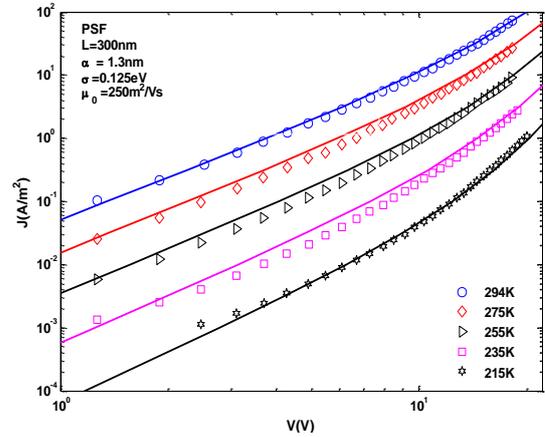


Fig. 1. Temperature dependent  $J-V$  characteristics of a PSF hole-only device with a layer thickness of 300 nm. Symbols are the experimental results from Ref. [15]. Lines are the numerically calculated results based on the IEGDM (color online)

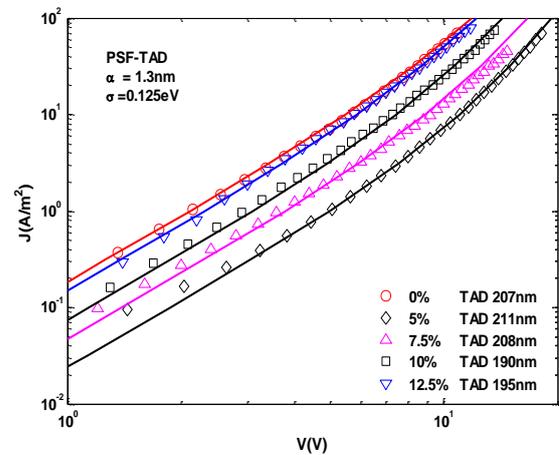


Fig. 2.  $J-V$  characteristics of hole-only devices based on PSF with 0%, 5%, 7.5%, 10% and 12.5% TAD concentration. Symbols are the experimental results from Ref. [15]. Lines are the numerically calculated results based on the IEGDM (color online)

As a next step, we consider the question whether the ECDM can also describe the  $J-V$  characteristics of PSF hole-only devices with various temperatures and TAD concentrations as successfully as the IEGDM. When employing the ECDM, we address the question whether site-energy correlation with this specific correlation function is present in the PSF-TAD copolymers. Here, we re-analyse the experimental data from Ref. [15] for hole-only devices based on PSF-TAD copolymers by employing the ECDM. In Fig. 3 and Fig. 4, the temperature dependent and TAD concentration dependent  $J-V$  characteristics of PSF hole-only devices are displayed, respectively. A clear observation is that the temperature dependent and TAD concentration dependent  $J-V$  characteristics of PSF hole-only devices can also be described within the ECDM as successfully as the IEGDM. For the PSF polymers with 0% TAD at different temperatures, a single set of parameters  $a = 0.24$  nm,  $\sigma = 0.15$  eV, and  $\mu_0 = 320$  m<sup>2</sup>/Vs can well describe the hole transport. For the PSF-TAD copolymers with different TAD concentrations, the obtained disorder parameter  $\sigma$  and intersite distance  $a$  are 0.15 eV and 0.24, respectively, same as the parameters used in Fig. 3. As for the used parameter  $\mu_0$  in Fig. 4, it ranges from 320, 50, 90, 115 and 215 m<sup>2</sup>/Vs for 0%, 5%, 7.5%, 10% and 12.5 % TAD concentration, respectively. At low concentrations the amine units act as hole traps and reduce the hole current, whereas above a critical concentration, percolation can take place between the amine units and the hole transport will become governed by guest-to-guest hopping, leading to an increase of the mobility with increasing amine concentration. This is reflected in a change of the IEGDM and ECDM hole-transport parameter  $\mu_0$ , which gradually increase with increasing TAD concentration.

It can be seen from Figs. 1-4 that equally good descriptions of the temperature dependent and TAD concentration dependent  $J-V$  characteristics of hole-only devices based on PSF and PSF-TAD copolymers can be obtained within both the IEGDM and ECDM. The optimal fitting parameter set for each model is obtained as follows. For a given  $\sigma$ ,  $a$ , and  $\mu_0$  combination, we determine the fitting error between the experimental results and calculated  $J-V$  curves for each individual temperature and TAD concentration. The total error is calculated as a sum of mean square errors of the fits for all temperatures and TAD concentrations. By minimizing this total error, we obtain the optimal model parameter set. Within the IEGDM and ECDM, the carrier density and electric field dependence of the mobility depend only on shape of the Gaussian density of states (DOS), specified by  $\sigma$  and  $a$ . Varying the mobility in the zero-density and zero-field limit, specified by  $\mu_0(T)$ , gives rise to an overall vertical shift of the  $J-V$  curves, but does not affect the shape. It can be clearly observed that within the IEGDM and ECDM the mobility at any temperature and TAD concentration is described by using

only three parameters, viz.  $\sigma$ ,  $a$ , and  $\mu_0$ , each parameter has a clear physical meaning. For the IEGDM, the values of  $\sigma$  and  $a$  obtained in this work are 0.125 eV and 1.3 nm, and for the ECDM  $\sigma = 0.15$  eV and  $a = 0.24$  nm. By optimizing the  $J-V$  curves position, the  $\mu_0$  can be determined for each individual temperature and TAD concentration within two models by using a shift along the vertical axes.

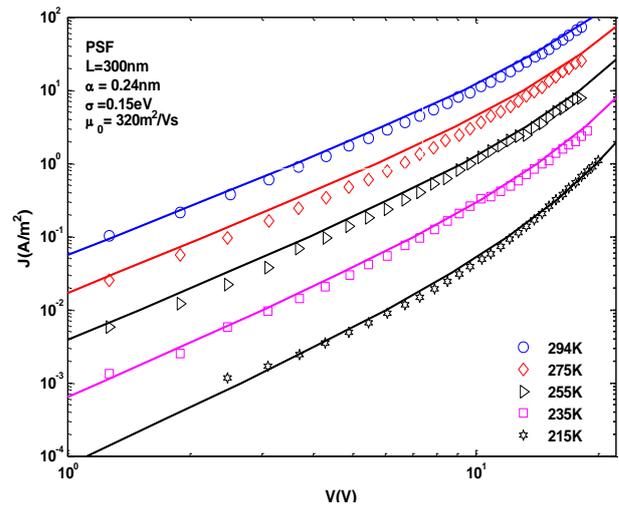


Fig. 3. Temperature dependent  $J-V$  characteristics of a PSF hole-only device with a layer thickness of 300 nm. Symbols are the experimental results from Ref. [15]. Lines are the numerically calculated results based on the ECDM (color online)

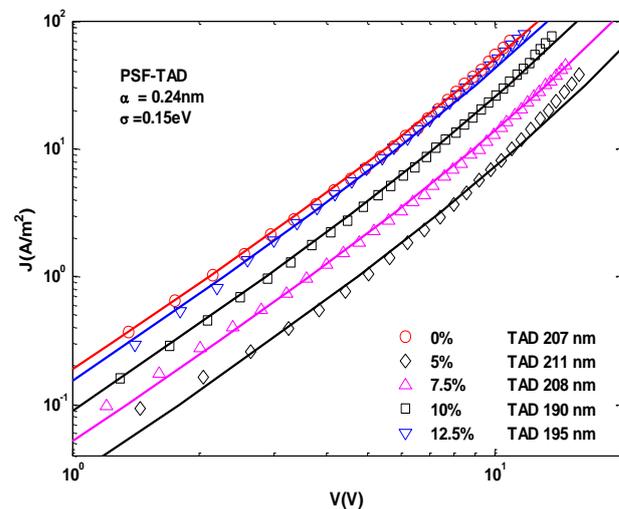


Fig. 4.  $J-V$  characteristics of hole-only devices based on PSF with 0%, 5%, 7.5%, 10% and 12.5% TAD concentration. Symbols are the experimental results from Ref. [15]. Lines are the numerically calculated results based on the ECDM (color online)

It should be noted that the optimal fit values of  $a$  as obtained from the IEGDM and ECDM are very different, viz. 1.3 nm and 0.24 nm, respectively. The value of  $a$  found for the IEGDM is very close to the typical value of organic semiconductors [13, 23], and

slightly smaller than the result reported by Nicolai et al. for PSF and PSF-TAD copolymers [14, 15]. However, the value of  $a$  found in the ECDM may be considered as unrealistically small for conjugated polymers. It is well known that intra-chain hopping between the rather long conjugated segments in conjugated polymers is also expected to be associated with a larger value of  $a$ . The distance between two subsequent monomer units is approximately 0.7 nm, and the conjugation length is believed to be at least five monomer units. These results indicate that in PSF and PSF-TAD copolymers studied correlation between the site energies is absent or insignificant. The values obtained for  $\sigma$  does not change this point of view. For disordered organic semiconductors, the Gaussian density of states  $\sigma$  is typically observed to fall in the range 0.06-0.16 eV, the optimal values of  $\sigma$  obtained within both models (0.125 eV for the IEGDM and 0.15 eV for the ECDM) are physically realistic. Owing to lack of independent experimental measurements on the width of the DOS, the value of  $\sigma$  cannot presently be applied to distinguish between the IEGDM and ECDM.

#### 4. Summary and conclusions

In conclusion, the hole transport and spatial correlation between the site energies in a series of PSF-TAD copolymers are studied. It is shown that the equally good descriptions of  $J-V$  characteristics of PSF hole-only devices with various temperatures and TAD concentrations can be obtained using both the IEGDM and ECDM, within which spatial correlation between the site energies is absent and is included, respectively. The width of the DOS  $\sigma$  and intersite distance  $a$  obtained in this work are found to be essentially independent of the TAD concentration within both the IEGDM and ECDM. For the IEGDM, the  $\sigma$  and  $a$  are 0.125 eV and 1.3 nm, and for the ECDM  $\sigma = 0.15$  eV and  $a = 0.24$  nm. The value of  $a$  obtained in the IEGDM is very close to the typical value of organic semiconductors, whereas the value of  $a$  obtained in the ECDM may be considered as unrealistically small for conjugated polymers. We view this as an indication that for the PSF-TAD copolymers studied correlation between the site energies is absent or plays a minor role. These results suggest that a successful analysis of the  $J-V$  curves using either model does not yet convincingly prove that the disorder is completely random or correlated, and the IEGDM can provide the appropriate framework for describing the guest concentration dependence of the charge transport in host-guest systems.

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