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Charge transport in a blue-emitting alternating block copolymer with a small spacer to conjugated segment length ratio

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We analyze current versus voltage data obtained using single carrier injection in several metal/polymer/metal sandwich structures. The polymer used in each case is a soluble blue-emitting alternating block copolymer. Our experimental results demonstrate that the electron transport is space-charge limited by the high density of traps having an exponential energy distribution (temperature dependent characteristic energy) in the copolymer. The electron mobility of $8 \times 10^{-10} \text{ cm}^2/\text{V s}$ is directly determined using space-charge-limited current analytical expressions. Hole transport is also space-charge limited, with a mobility of $2 \times 10^{-6} \text{ cm}^2/\text{V s}$. A hole trap with energy 0.17 eV is observed. We compare these results with those obtained for related block copolymers with different spacer and conjugated segment lengths and discuss the influence of spacer length and conjugated segment length on the charge transport properties. © 2000 American Institute of Physics. [S0021-8979(00)04501-1]

I. INTRODUCTION

Charge carrier mobility is a fundamental parameter in characterizing the performance of pristine semiconductor polymers, which are used *inter alia* as electroluminescent materials in light-emitting diodes (LEDs). To enhance quantum efficiency and response time, and to reduce operating voltage, high charge carrier mobility is required.¹ Charge carrier mobilities have an important role in achieving a balanced injection of positive and negative charge carriers and the spatial location of the emissive region in organic LEDs.^{2,3}

Conjugated oligomers have well defined optical emission and absorption characteristics and these polymers also present interesting mechanical properties. A strategy that permits emission control in a way similar to the variation of oligomer molecular mass, but without loss of the useful mechanical properties of polymers, is the use of block copolymers with conjugated and nonconjugated (spacer) segments. The emission color control is in this case achieved by control of the length of the conjugated segment.^{4,5}

When carrier traps are present in a polymer and electron and hole transport in the polymer shows space-charge-limited characteristics, the charge carrier mobility can be readily determined by space-charge-limited current measurements. Recent studies have demonstrated that electron and

hole transport in several electroluminescent organic polymers is in fact space-charge limited.^{6–11}

In this article we investigate the charge transport characteristics of a electroluminescent block copolymer, poly(1,4-butanenedioxy-2,6-dimethoxy-1,4-phenylene-1,2-ethynylene-1,4-phenylene-1,2-ethynylene-1,4-phenylene-1,2-ethynylene-3,5-dimethoxy-1,4-phenylene) (polymer I in Fig. 1) and compare its transport characteristics including mobilities and trap distribution energies with recently investigated related copolymers (polymers II and III in Fig. 1),^{9–11} which have different spacer lengths and/or different conjugated segment lengths. This comparison permits a discussion of the role of the spacer and the role of the conjugated segment length in the transport characteristics of these copolymers. The experimental procedure and the strategy used to determine the charge carrier mobility in this work are identical to that used for copolymers II and III (Refs. 9–11) in order to permit a direct comparison of results.

II. SPACE-CHARGE-LIMITED CONDUCTION

The mobility of the charge carriers can be extracted from current versus voltage curves of a polymer film sandwiched between two conducting electrodes. When the transport in a trap-free conjugated polymer is not injection limited, i.e., the current is limited due to space-charge accumulation, the current density j is expressed by

$$j = \frac{9}{8} \epsilon \mu \frac{V^2}{d^3}, \quad (1)$$

where ϵ is the permittivity of the polymer, μ is the charge

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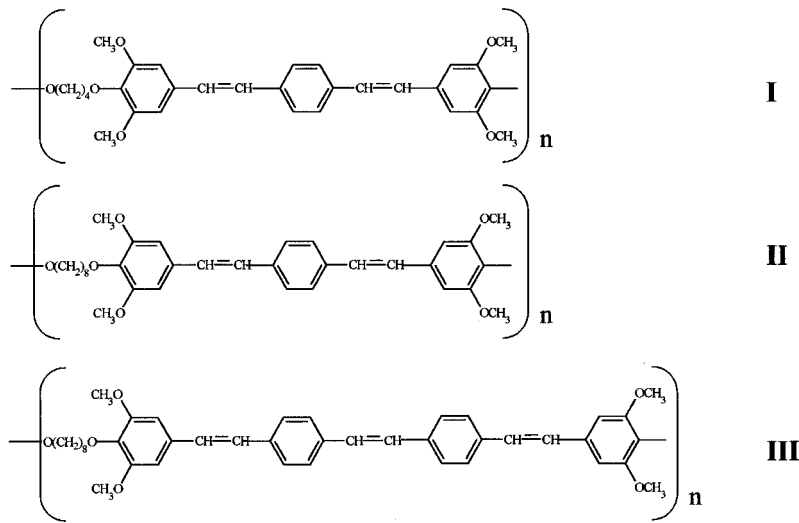


FIG. 1. Structure of (I) poly(1,4-butanedioxy-2,6-dimethoxy-1,4-phenylene-1,2-ethynylene-1,4-phenylene-1,2-ethynylene-3,5-dimethoxy-1,4-phenylene); (II) poly(1,8-octanedioxy-2-dimethoxy-1,4-phenylene-1,2-ethynylene-1,4-phenylene-1,2-ethynylene-3,5-dimethoxy-1,4-phenylene); and (III) poly(1,8-octanedioxy-2,6-dimethoxy-1,4-phenylene-1,2-ethynylene-1,4-phenylene-1,2-ethynylene-3,5-dimethoxy-1,4-phenylene).

carrier mobility, and d is the polymer film thickness. This charge transport case is characterized by $j(V, d)$ dependence expressed by the above equation and by a weak $\mu(T)$ dependence. If traps present in the polymer layer are located at a single discrete energy level, this expression is modified to¹²

$$j = \frac{9}{8} \varepsilon \theta \mu \frac{V^2}{d^3}, \quad (2)$$

where

$$\theta = \frac{n}{n + n_t} \text{ or } \theta = \frac{p}{p + p_t} \quad (3)$$

for electrons and holes, respectively (n is the density of free electrons, n_t is the density of trapped electrons, p is the density of free holes and p_t is the density of trapped holes). In this case, $\theta \mu \equiv \mu_{\text{eff}}$ is the effective charge carrier mobility. To distinguish between μ and μ_{eff} a more detailed investigation, including, for example, temperature dependence of the mobility and dependence on the electrode work function must be performed.^{10,11} The parameters p and p_t (considering specifically the case of hole traps) are given by

$$p = N_v \exp\left(-\frac{E_f - E_v}{\kappa T}\right) \quad (4)$$

and

$$p_t = \frac{N_t}{1 + \exp\left(\frac{E_f - E_t}{\kappa T}\right)}, \quad (5)$$

where N_v is the density of states in the valence band, N_t is the density of traps, E_f is the relevant Fermi level, E_v is mobility edge energy for positive charge and E_t is the energy level of the traps; κ is the Boltzmann constant.

Therefore, for traps lying at an energy E_t below the E_f such that $(E_f - E_t)/\kappa T > 1$ and $p_t > p$, we have

$$\theta = \frac{N_v}{N_t} \exp\left(-\frac{E_t - E_v}{\kappa T}\right). \quad (6)$$

Substituting for θ yields

$$j = \frac{9}{8} \varepsilon \mu \frac{N_v}{N_t} \exp\left(-\frac{E_t - E_v}{\kappa T}\right) \frac{V^2}{d^3}, \quad (7)$$

so that the effective mobility can be written as

$$\mu_{\text{eff}} = \mu \frac{N_v}{N_t} \exp\left(-\frac{E_t - E_v}{\kappa T}\right), \quad (8)$$

yielding a strong temperature dependence for μ_{eff} .

In amorphous materials a single discrete energy level may no longer be a reasonable approximation. In this case, the localized states (trap) density is expected to be in the form of an exponential distribution at energies E within the forbidden energy gap, such that

$$h(E) = \frac{N_t}{E_c} \exp\left(-\frac{E}{E_c}\right), \quad (9)$$

where N_t is the trap center density and E_c is the characteristic energy of the distribution. The $j(V, d)$ dependence in this case is of the form¹²

$$j \propto \frac{V^{m+1}}{d^{2m+1}}, \quad (10)$$

where $m \equiv T_c/T$; $E_c \equiv \kappa T_c$ (T is the absolute temperature and T_c is the characteristic temperature).

In all the cases described above, μ is considered to be independent of the electric field strength although in some cases field dependent mobilities were also reported for several organic compounds.^{13,14}

III. EXPERIMENT

The metal/polymer/metal devices were prepared on a glass substrate. The bottom metal electrode was evaporated onto the glass and the polymer was spin coated in CHCl_3 solution (5 mg/ml) onto this electrode. The top metal electrode was then evaporated onto the polymer layer. The metals used were Al, Cu and Au, with work functions (ϕ) of 4.28, 4.65 and 5.1 eV, respectively. The polymer film thickness was measured using a surface profiler. The current-voltage $I(V)$ measurements were made by increasing the

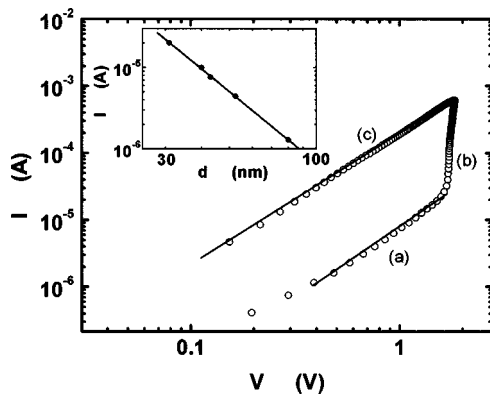


FIG. 2. $I(V)$ characteristics of a Cu/copolymer-I/Al device ($d=43$ nm). Data obtained for Cu were positively biased. In region (a) $j \propto V^2$ (increasing applied voltage). Region (b) corresponds to trap filling. Region (c) follows $j \propto V^2$ (decreasing applied voltage). The inset presents the dependence of current on the copolymer-I layer thickness at constant $V=1$ V.

voltage from zero to a maximum value at a rate of 0.1 V s^{-1} and then decreasing it to zero at the same rate. Measurements were carried out at room temperature, except when the temperature dependence was determined. Further details concerning sample geometry were as reported earlier.¹⁵ The dielectric constant of the polymer was assumed to be 3.

IV. RESULTS

The $j(V)$ characteristics of a Cu/copolymer-I/Al device (Fig. 2) show three different regions. At increasing voltage, $j \propto V^2$ [region (a)] up to the trap filling region [region (b)]. After the maximum applied voltage value is achieved, the current density with decreasing applied voltage follows $j \propto V^2$ [region (c)]. The $j(V)$ characteristics of the Cu/copolymer-I/Al device were measured for different copolymer-I layer thickness. In the inset of Fig. 2 we present the current dependence on copolymer-I layer thickness, for $V=1$ V. A linear fit to these data gives $\partial \log I / \partial \log d = -2.91$.

Using Eq. (2) we determined the value of μ_{eff} for regions (a) and (c) of Fig. 2. We obtained $\mu_{\text{eff}} = (6.4 \pm 1.0) \times 10^{-8} \text{ cm}^2/\text{V s}$ and $\mu_{\text{eff}} = (1.9 \pm 0.9) \times 10^{-6} \text{ cm}^2/\text{V s}$, respectively.

The temperature dependence of μ_{eff} is presented in Fig. 3. It corresponds to segment (a) of the $j(V)$ curve of Cu/copolymer-I/Al devices (see also Fig. 2). The linear fit to the data of Fig. 3 results in $\partial \ln \mu / \partial T^{-1} = -2004 \text{ K}^{-1}$, which corresponds to $E_t - E_v = 0.17$ [see Eq. (8)].

We also characterized Au/copolymer-I/Al devices (Fig. 4) which present $j(V)$ characteristics with a different structure from those observed for Cu/copolymer-I/Al devices. A region of the $j(V)$ curve observed at increasing voltage follows $j \propto V^2$. Using Eq. (2), $\mu_{\text{eff}} = (3.2 \pm 1.1) \times 10^{-8} \text{ cm}^2/\text{V s}$ was determined, a value quite similar to that observed for Cu/copolymer-I/Al devices for increasing voltage [region (a) of Fig. 2]. The Au/copolymer-I/Al device shows a region at higher voltages in which the current increase is stronger than $j \propto V^2$. This region could not be investigated in detail, because devices constructed with Au could not withstand high applied voltage. This phenomenon was also observed in

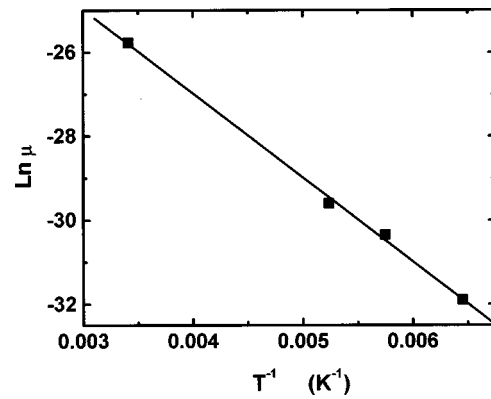


FIG. 3. Temperature dependence of μ_{eff} for a Cu/copolymer-I/Al device ($d=42$ nm). The segment (a) of $j(V)$ curves seen in Fig. 2 was used to calculate the mobility.

other electroluminescent block copolymers.^{9,10} Using Au/copolymer-I/Al devices with different copolymer-I layer thicknesses the $I(d)$ dependence at $V=1$ V presented in the inset of Fig. 4 was determined. These data yield $\partial \log I / \partial \log d = -2.99$.

The $I(V)$ characteristics of Al/copolymer-I/Al devices are shown in Fig. 5. For increasing voltage, the curve can again be divided into three regions. Region (a) corresponds to ohmic injection ($j \propto V$). Region (b) corresponds to $j \propto V^{m+1}$ ($m \geq 1$) while region (c), to $j \propto V^2$. From measurements of the $I(V)$ characteristics at different temperatures we obtained the values of m at different temperatures and calculated the corresponding E_c value. These results are presented in Table I.

In the inset of Fig. 5 we show the dependence of applied voltage on copolymer-I layer thickness, in the form $I(d^2)$, for Al/copolymer-I/Al devices, at a fixed current of $7 \times 10^{-8} \text{ A}$ [corresponding to region (b)]. For this case when $m \approx 2$ a nearly linear dependence of V on d^2 is expected [see Eq. (10)].

Using Eq. (2), $\mu_{\text{eff}} = (7.8 \pm 1.1) \times 10^{-10} \text{ cm}^2/\text{V s}$ is obtained for the region $j \propto V^2$ [region (c)].

The $I(V)$ dependence of an Al/copolymer-I/Al device on temperature is presented in Fig. 6. The $I(V)$ curve is tem-

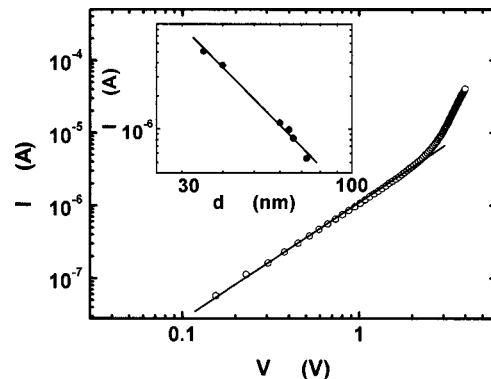


FIG. 4. $I(V)$ characteristics of an Au/copolymer-I/Al device ($d=64$ nm). The $j(V)$ characteristics of the device are characterized by a region in which $j \propto V^2$, observed at increasing applied voltage. Inset: $I(d)$ dependence at $V=1$ V.

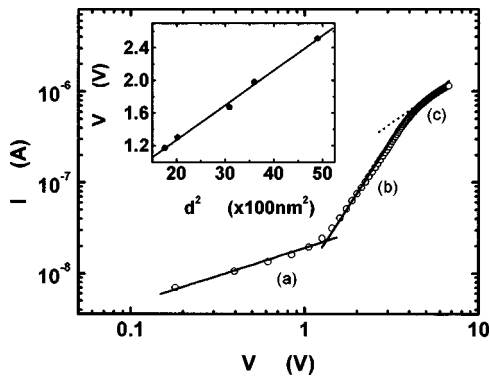


FIG. 5. $I(V)$ characteristics of an Al/copolymer-I/Al device ($d=56$ nm). Region (a) corresponds to ohmic injection ($j \propto V$). Region (b) corresponds to $j \propto V^{m+1}$ and region (c) to $j \propto V^2$. Inset: Dependence of the applied voltage on copolymer-I layer thickness at a fixed current of 7×10^{-8} A.

perature independent in the region corresponding to $j \propto V^2$. For this reason and to avoid possible sample degradation, we limited the applied voltage to the value corresponding to the beginning of the $j \propto V^2$ region.

V. DISCUSSION

The $I(V, d)$ dependence observed in X/copolymer-I/Al (X: Cu, Au and Al) devices demonstrates that the charge carrier transport in copolymer-I is space-charge limited for both electrons and holes.

Based on the electrode work function values and the electron affinity expected for these copolymers,⁹ we conclude that Cu/copolymer-I/Al devices operate in a hole-only mode (confirmed with Au/copolymer-I/Al devices). The $I(V)$ characteristics of Cu/copolymer-I/Al devices follow a $j \propto V^2$ regime, defining a trap filling region. It is observed that the effective hole mobility is increased after the trap filling process, indicating that the quasicchemical potential exceeds the trap level leading to $n \gg n_t$, so that $\theta \approx 1$ [see Eq. (3)]. Under these conditions, the effective charge carrier mobility equals the free charge carrier mobility. The value for hole mobility is then obtained at decreasing applied voltage. Furthermore, $\partial \log I / \partial \log d \approx -3$ [see Eq. (2)], so that the $j(V, d)$ characteristics unequivocally demonstrate that the charge transport is, in this case, space-charge limited.

The temperature dependence of the mobility determined from segment (a), Fig. 2, which is presented in Fig. 3, is consistent with the existence of a trap level [see Eq. (8)]. These data permit the location of the trap level at 0.17 eV above the hole mobility edge. It is important to observe that the μ_{eff} observed in Cu/copolymer-I/Al devices before trap

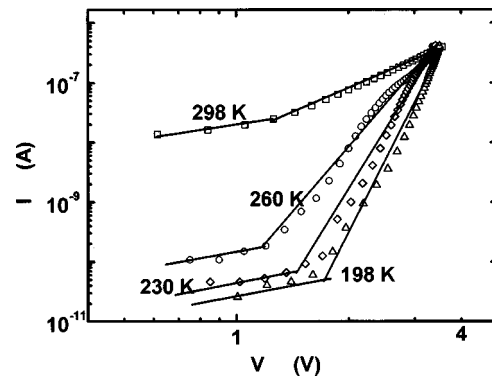


FIG. 6. Dependence of the $I(V)$ characteristics of an Al/copolymer-I/Al device ($d=56$ nm) on temperature.

filling is quite similar to that observed in Au/copolymer-I/Al devices. This result indicates that these traps are also not filled in the $I(V)$ measurements carried out with Au/copolymer-I/Al devices. Also in this case, the relation $\partial \log I / \partial \log d \approx -3$ expected from Eq. (2) is fulfilled.

The high applied voltage region of the $I(V)$ characteristics of Au/copolymer-I/Al could not be investigated due to device degradation, so that the origin of the increase in the $\partial \ln I / \partial \ln V$ value could not be identified. Our results do not provide a definitive explanation of this phenomenon, but three factors may be considered. One is the simultaneous injection of minority charge carriers (in this case, electrons). When majority charge carriers are injected, minority charge carrier injection by tunneling is facilitated.¹⁶ The second possible source of this effect is that the maximum achieved applied voltages for these devices correspond to trap filling. At high field strengths, a field dependent mobility also implies in such an increase.¹⁷⁻¹⁹

The $I(V, d)$ dependence observed in Al/copolymer-I/Al devices demonstrates that the transport is space-charge limited and that the traps are energetically distributed following an exponential distribution [see Eqs. (9) and (10)]. In copolymer-I, in contrast to what has been previously observed in copolymers II and III, E_c is temperature dependent (Table I). The value of E_c is generally related to order in the material: a higher E_c indicates a higher degree of disorder, with more localized state levels located deep within the energy gap. At room temperature, the E_c value for copolymer-I (0.05 eV) is nearly the same as that found for copolymer-II (0.06 eV) and for copolymer-III (0.05 eV).⁹ This demonstrates that at room temperature the spacer length of copolymer-I is enough to accommodate forces that would be responsible for energetic disorder. At lower temperatures well below the observed T_g ,²⁰ the value of E_c increases, indicating that the spacer loses this capability. At lower temperatures, the E_c value observed in copolymer-I is higher than that observed in nominal homopolymers such as PPV.^{21,22}

The Al/copolymer-I/Al devices show a temperature dependence of the current (and consequently of the mobility) up to a limit at which the current becomes temperature independent. This limit corresponds to the voltage where the $j(V)$ characteristics transit from the regime governed by Eq.

TABLE I. Characteristic energy E_c measured at different temperatures for an Al/copolymer-I/Al device.

T (K)	E_c (eV)
198	0.17
230	0.18
260	0.13
298	0.05

(10) to the regime governed by Eq. (1) or (2) with $\theta \approx 1$ [which corresponds to the transition from region (b) to region (c) in Fig. 5, or to the maximum value of V in Fig. 6]. This indicates that the traps are filled and that the measured mobility is that of the corresponding free charge carrier. Considering that the value obtained for Al/copolymer-I/Al devices is lower than that observed for holes, we attribute this mobility value to the negative charge carriers.

VI. CONCLUSIONS

Current versus voltage measurements have been made using single carrier injection into metal/polymer/metal structures. The polymer is a soluble blue-emitting alternating block copolymer (copolymer-I, Fig. 1). Our experimental results demonstrate that the electron transport is space-charge limited by a large number of traps with an exponential energy distribution and a temperature dependent characteristic energy. The electron mobility of $8 \times 10^{-10} \text{ cm}^2/\text{V s}$ is directly determined using space-charge-limited current analytical expressions. The hole transport is also space-charge limited, with a mobility of $2 \times 10^{-6} \text{ cm}^2/\text{V s}$, which can be significantly reduced by a trap with energy 0.17 eV, depending on the measurement conditions.

The hole mobility observed in polymer-I ($2 \times 10^{-6} \text{ cm}^2/\text{V s}$) is significantly higher than that reported for polymer-II ($4 \times 10^{-7} \text{ cm}^2/\text{V s}$).⁹ This result is consistent with the fact that a reduction in the spacer length implies a reduction in the average tunneling distance, making charge transport by hopping more facile. In polymer-III a hole mobility of $2 \times 10^{-6} \text{ cm}^2/\text{V s}$ has been reported.¹⁰ The mobility trend from copolymer-II to copolymer-III has been tentatively explained as being due to a higher conjugated material to spacer material volume ratio for copolymer-III,⁹ which at constant spacer length, reduces the average tunneling distance when longer conjugated segments occur.⁹ The fact that polymers I and III gave the same hole mobility value could imply that a longer conjugated segment and shorter aliphatic spacer may lead to higher hole mobility.

The electron mobility of copolymer-I was determined to be $8 \times 10^{-10} \text{ cm}^2/\text{V s}$. This value is only slightly larger than that reported for copolymer-II ($5 \times 10^{-10} \text{ cm}^2/\text{V s}$). Copolymer-I has a reduced spacer length compared to copolymer-II. This characteristic, together with the fact that copolymer-I has a lower E_c than copolymer-II (0.05 and 0.06

eV, respectively) makes it reasonable to expect a higher electron mobility in copolymer-I than in copolymer-II, which was experimentally confirmed.

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