Field-Effect Mobility of Charge Carriers in Blends of Regioregular Poly(3-alkylthiophene)s

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Thin film transistors based on regioregular poly(3-hexylthiophene) (PHT) and poly(3-decylthiophene) (PDT) and their binary blends were investigated and the blend composition dependence of the charge carrier mobility was determined. The field-effect mobility of holes (μ_h) in a series of 10 binary blends of poly(3-hexylthiophene) ($\mu_h = 0.02$ cm^2/(V s)) and poly(3-decylthiophene) ($\mu_h = 8 \times 10^{-5}$ cm^2/(V s)) was found to be relatively high (2×10^{-3} cm^2/(V s)) and constant over a broad composition range (5−80 wt % PDT). Above 80% PDT, the hole mobility decreased exponentially with composition. Atomic force microscopy of the homopolymers and blends confirmed the absence of phase separation in the blends. A similar interlayer d spacing that is intermediate between those of the homopolymers is implied by the constant hole mobility in the single-phase crystalline blends of 5−80 wt % PDT. These results demonstrate that blends of conjugated polymers are alloy semiconductors in which high and tunable charge carrier mobility can be realized.

Introduction

Charge transport in conjugated polymer semiconductors is of broad technological and fundamental interest because of the growing applications of the materials in thin film transistors,[1−9] light emitting devices,[10] photovoltaic cells,[11] photodetectors,[12,13] and electrophotographic imaging.[13] Among conjugated polymer semiconductors, charge carrier transport has been most extensively investigated in the poly(3-alkylthiophene)s.[3−8] An early study of electrochemically synthesized thin films of poly-(thiophene-2, 5-diyl) gave a field-effect hole mobility of about 10^{-5} cm^2/(V s).[13] A study of spin coated thin films of chemically synthesized regiorandom poly(3-alkylthiophene)s showed field-effect mobilities of 6×10^{-7} to 2×10^{-4} cm^2/(V s) depending on the alkyl chain length: 10^{-4} cm^2/(V s) for poly(3-hexylth-iophene) and 6×10^{-7} cm^2/(V s) for poly(3-decylthiophene), for example.[4a] Advances in the synthesis of regioregular poly-(3-alkylthiophene)s have resulted in the achievement of significantly higher hole mobilities in this class of conjugated polymer semiconductors.[14] Field-effect hole mobilities as high as 0.05 to 0.1 cm^2/(V s) have been measured in solution cast regioregular poly(3-hexylthiophene).[5−7] A large decrease in mobility with increasing length of the alkyl side chain length has also been observed in a series of regioregular poly(3-alkylthiophene)s.[1,5,8] The field-effect hole mobility varied from 10^{-2} and 2.9×10^{-3} cm^2/(V s) in poly(3-butylthiophene) and poly(3-hexylthiophene), respectively, to 5×10^{-6} cm^2/(V s) in poly(3-octadecylthiophene).[8]

Although there has been significant interest in the structural, thermochromic, and light emitting properties of blends of polythiophenes[15,16] and other conjugated polymers,[17,18] the charge carrier mobility in such blends has heretofore not been investigated or reported. It was shown by X-ray diffraction studies of binary blends of regioregular poly(3-alkylthiophene)s that the materials formed single-phase crystalline structures with an interlayer d spacing that was intermediate between those of the component homopolymers.[15a] We recently proposed that blends of conjugated polymers represent a new class of homologous series of molecular semiconductors that can be used to

investigate morphology-charge transport relationships.[9b] The first thin film transistors based on blends of conjugated polymers were demonstrated and used to determine the composition-dependent field-effect mobility of *electrons* in the blends.[9b] Here we focus on blends of regioregular poly(3-alkylthiophene)s that provide an opportunity to study the blend composition dependence of the field-effect mobility of *holes* in the alloy semiconductors. Blends of conjugated polymers have proven effective in improving or optimizing electronic, optoelectronic, and photonic properties such as enhancement in electroluminescence,[16,17] enhancement of photovoltaic properties,[11a,d] photoinduced charge transfer,[18a] and bipolar conductivity.[18b] Conjugated polymer blends also provide the possibility of a continuous tunability of morphology and charge transport in conjugated polymer-based alloy semiconductors.

In this paper, we report the fabrication and investigation of charge carrier transport in thin film transistors based on a series of binary blends of regioregular poly(3-hexylthiophene) and poly(3-decylthiophene). The blend composition dependence of the field-effect mobility of holes was determined and found to have an unusual feature. A relatively high and constant hole mobility of about 2×10^{-3} cm^2/(V s) was observed over a broad composition range (5−80 wt % PDT) whereas an exponential decrease of the mobility with composition was observed above 80 wt % PDT. Atomic force microscopy of the homopolymers and binary blends was used to confirm the homogeneous single-phase nature of the PHT/PDT blends.

Experimental Section

Materials. Regioregular poly(3-hexylthiophene) and poly-(3-decylthiophene) with head-to-tail (HT) coupling exceeding 98.5% were purchased from Aldrich and Rieke Metals Inc. (Lincoln, NE), respectively, and used without further purification. The synthesis of these polymers has been reported.[14c] High-purity HPLC grade chloroform was used as received.

Binary blends of poly(3-hexylthiophene) and poly(3-decylthiophene) were prepared by dissolving binary mixtures of PHT and PDT in chloroform with resulting solutions of 0.1 wt % of
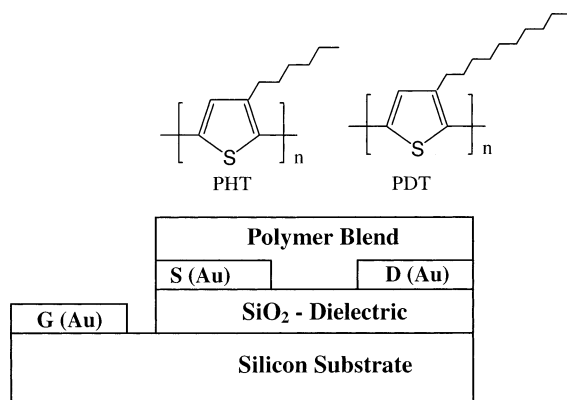
Figure 1. Molecular structures of regioregular poly(3-alkylthiophene)s (PHT, PDT) and a schematic of the thin film transistor.

total polymer. Composition of blends in this paper refers to weight percentage of PDT and 5, 10, 20, 40, 50, 60, 80, 85, 90, and 95 wt. % PDT blends were prepared and investigated. All thin films of the homopolymers and blends were spin coated from the chloroform solutions at 1200 rpm for 30 s. The films were dried overnight (10–12 h) at 60 °C in a vacuum oven to remove any residual solvent. Blend thin films (20–30 nm thick) spin casted on glass slides were homogeneous and they showed good optical transparency.

Atomic Force Microscopy of Polymer Thin Films. The morphology of PHT and PDT homopolymers and their blends was studied using atomic force microscopy (AFM) (Digital Instruments, Santa Barbara, CA) in standard tapping mode. The polymer thin films were spin coated from their chloroform solutions onto Si substrates having a 300-nm SiO_2 surface layer to emulate the device structure. The film thickness of the polymer samples used for AFM imaging was 20–30 nm. The film thickness was measured by an Alpha-Step 500 profilometer (KLA Tencor, Mountain View, CA) with an accuracy of 1 nm. Topographic and phase images were taken.

Fabrication and Characterization of Thin Film Transistors. The thin film field-effect transistors were fabricated by using a bottom contact geometry shown in Figure 1. Heavily doped Si with a conductivity of 10^3 S/cm was used as a gate electrode with a 300-nm thick SiO_2 layer as the gate dielectric. By means of standard photolithography and a vacuum sputtering system (2×10^{-6} Torr), two 90-nm thick gold electrodes (source and drain) with a 10-nm thick TiW alloy adhesive layer were fabricated onto the SiO_2/Si substrate.[9] The channel length (L) was in the range of 20–50 μm and the channel width (W) was in the range 400–1500 μm. A gold contact pad was also deposited onto the gate electrode to make ohmic contact. Finally, on top of this device structure, thin films (20–30 nm) of poly-(3-alkylthiophene) or a binary PHT/PDT blend were spin coated from a chloroform solution and dried overnight (10–12 h) at 60 °C in a vacuum oven. Thermal annealing of all the FETs at 100 °C for 5 min generally improved the off state characteristics and thus the on/off current ratio. Electrical characteristics of these devices were measured at room temperature using an HP 4155A semiconductor parameter analyzer (Yokogawa Hewlett-Packard, Tokyo). All the fabrication and measurements were done under ambient laboratory conditions.

Results and Discussion

Morphology of Homopolymer and Blend Thin Films. Figure 2 shows the AFM topographic and phase images of a PHT thin film on SiO_2/Si substrate. Cylindrical rodlike crystallites with about 20–30 nm in diameter and lengths ranging
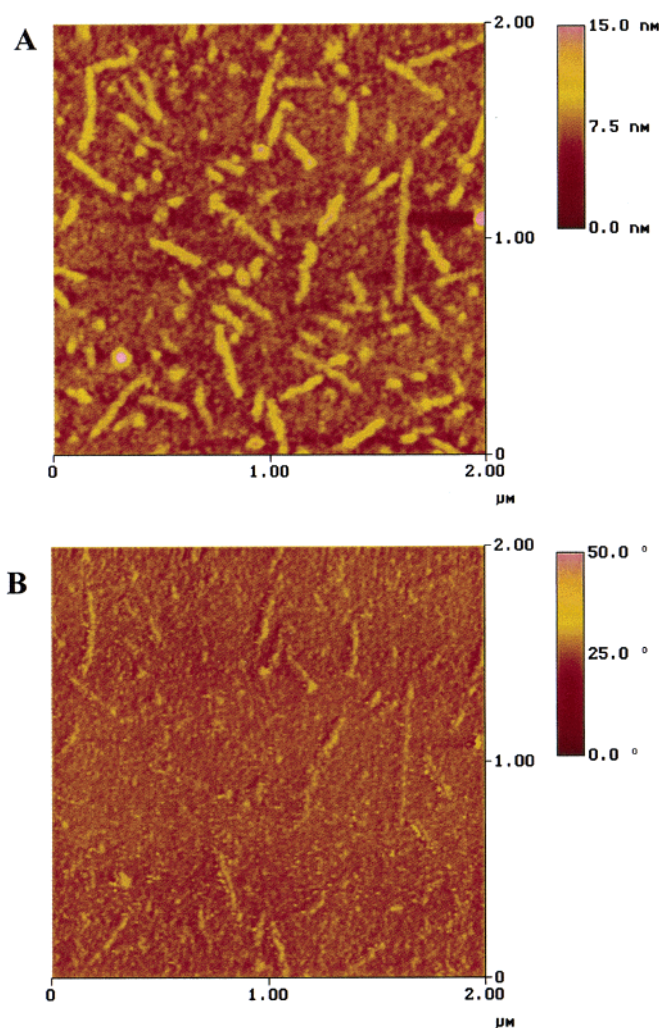

Figure 2. AFM tapping mode topographical (A) and phase (B) images of a PHT thin film on SiO_2 surface on a silicon substrate.

from 250 nm to 1 μm are clearly evident especially in the topographic image. These randomly oriented nanorod crystallites are formed in the solid state after evaporation of the solvent and not during the spin coating process. The lack of any preferred orientation of the nanorod crystallites is a consequence of the radially isotropic spin coating process that was used to prepare the films.[19] PHT thin films that are solution cast on pretreated surfaces have been shown to exhibit preferred alignment of crystalline lamellae parallel or perpendicular to the substrate surface.[6b] The nanorod geometry of the PHT crystallites in the present films is likely a consequence of the hydrophilic SiO_2 surface on which the thin films were deposited. The AFM topographic and phase images of a PDT thin film on SiO_2/Si substrate are shown in Figure 3. Large crystallites similar to those seen in PHT thin films were not observed in PDT thin films. However, these observations from Figures 2 and 3 do not imply that PHT is more crystalline than PDT because the degree of crystallinity does not scale linearly with crytallite size. In fact poly(3-alkylthiophene)s with longer alkyl side chains are often more crystalline that PHT.[15,20]

AFM study of the blend thin films was done with the goal of confirming either a homogeneous single-phase or a phase-separated morphology when compared to the homopolymers. AFM images of a 50% blend thin film on SiO_2/Si substrate are shown in Figure 4. A small number of nanorodlike crystallites are observed. AFM images of other blend compositions (10, 60, and 80 wt % PDT) showed a trend of decreasing nanorodlike
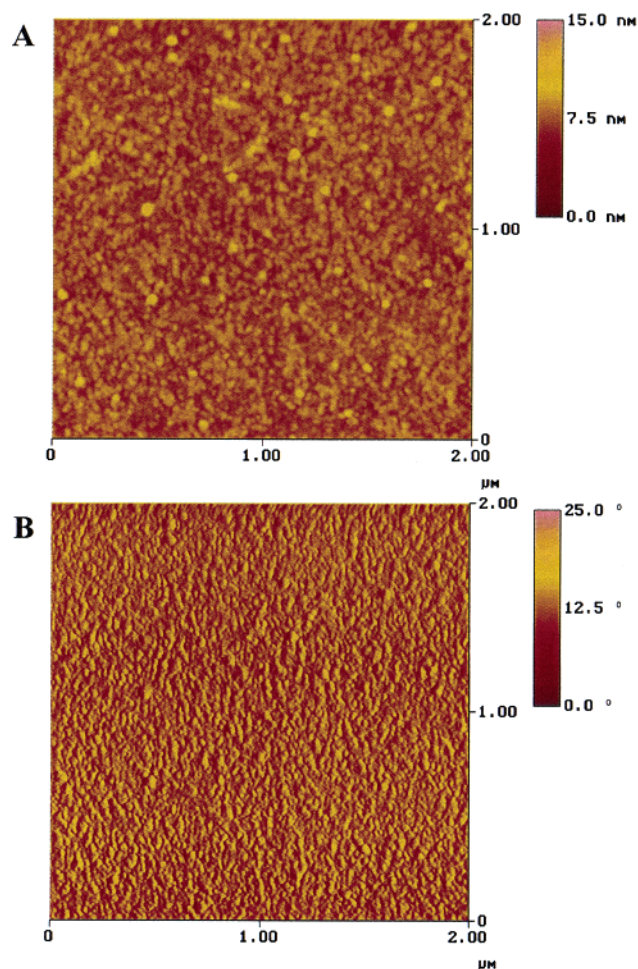
Figure 3. AFM tapping mode topographical (A) and phase (B) images of a PDT thin film on a SiO$_2$ surface on a silicon substrate.

crystallites with increasing concentration of PDT. A comparison of the blend thin film morphology with those of the homopolymers (PHT and PDT) and prior X-ray diffraction studies of blends of regioregular poly(3-alkylthiophene)s,[15a] shows that the blends of different compositions were single-phase crystalline materials in bulk. It must be noted that because the X-ray diffraction pattern does not show the peaks exclusive to the crystalline regions (nanorods), the possibility that those regions are exclusive to one blend component cannot be ruled out completely.

Electrical Characteristics of Thin Film Transistors. Figure 5 shows the output characteristics of an FET made from PHT homopolymer, showing the source-drain current (I_d) vs drain voltage (V_d) at different gate voltages (V_g). The observed electrical characteristics are typical $I-V$ curves for a p-channel FET operating in the accumulation mode. In the saturation region ($V_d > V_g - V_t$), I_d can be described by eq 1 where μ_h is the field-effect hole mobility, W is the channel width, L is the channel length and C_o is the capacitance per unit area of the gate dielectric layer (SiO$_2$, 300 nm, C_o = 11 nF/cm^2) and V_t is the threshold voltage.[1a,d,21] The field-effect mobility was calculated by plotting $I_d^{1/2}$ vs V_g (Figure 5B) and equating slope of this plot to $(W/2L)C_o\mu_h$.

$$I_d = (W/2L)\mu_h C_o(V_g - V_t)^2 \qquad (1)$$

The resulting hole mobility for the PHT FET with the $I-V$ characteristics shown in Figure 5A is 0.01 cm^2/(V s) and the maximum I_{on} ($V_g = -100$)/I_{off} ($V_g = +20$) ratio observed was

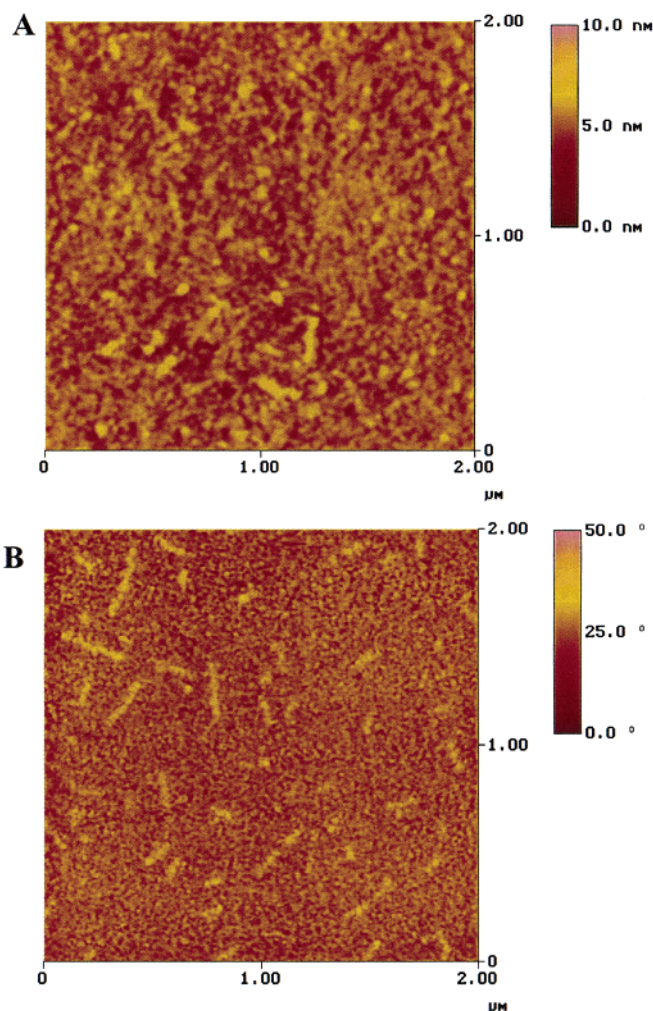**Figure 4.** AFM tapping mode topographical (A) and phase (B) images of a 50% PDT blend thin film on a SiO$_2$ surface on a silicon substrate.

10^4. With time after annealing the hole mobility increased to 0.02 cm^2/(V s) and the I_{on}/I_{off} ratio decreased to 10^2, suggesting that the conductivity of the film had increased. This value of the hole mobility in regioregular PHT is comparable to the highest values previously reported for this polymer.[5-7] We note that slightly higher mobilities in the range of 0.05 to 0.1 cm^2/(V s) have also been reported for this polymer when a special hydrophobic adhesion layer was used to improve the self-ordering of the layered crystalline PHT thin films on SiO$_2$/Si surfaces.[6] Such a surface treatment was not explored in the present study.

The output and transfer characteristics of a PDT FET are shown in Figure 6. The hole mobility calculated using eq 1 was 8 × 10^{-5} cm^2/(V s) with an I_{on}/I_{off} ratio of 3 × 10^3. This value of the hole mobility in regioregular PDT is a factor of 250 smaller than in regioregular PHT and this is in general agreement with prior observations on the trend of field-effect mobility with alkyl chain length.[1,4] However, our μ_h value for PDT is about an order of magnitude smaller than that recently reported (μ_h = 9 × 10^{-4} cm^2/(V s)) by Kaneto et al.[8] We believe that our μ_h value for PDT is more reliable than that reported by these authors. The μ_h reported by these authors for PHT was only 2.9 × 10^{-3} cm^2/(V s), giving rise to only a factor of 3.2 difference with PDT. The quality of the FETs reported by Kaneto et al. was poor, judging by the rather low on/off current ratio of only 15. The rather thick films (1-2 μm) used by these authors in fabricating their poly(3-alkylthiophene) FETs likely led to high conductivity and the low on/off current ratios.[8]
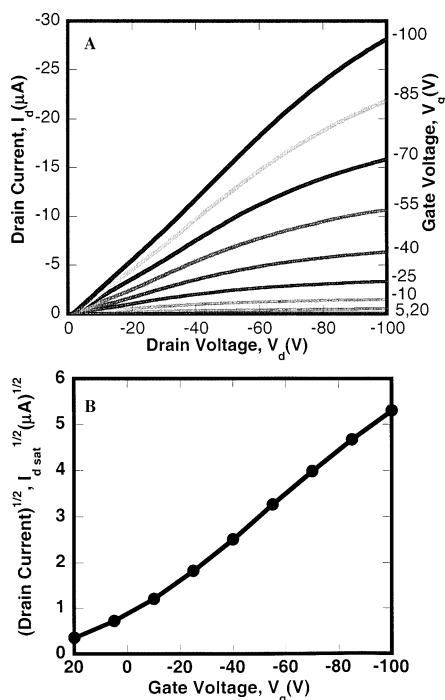
Figure 5. Output characteristics (A) and $I_d^{1/2}$ versus V_g plot (B) of a PHT FET with $W = 400$ μm and $L = 20$ μm.

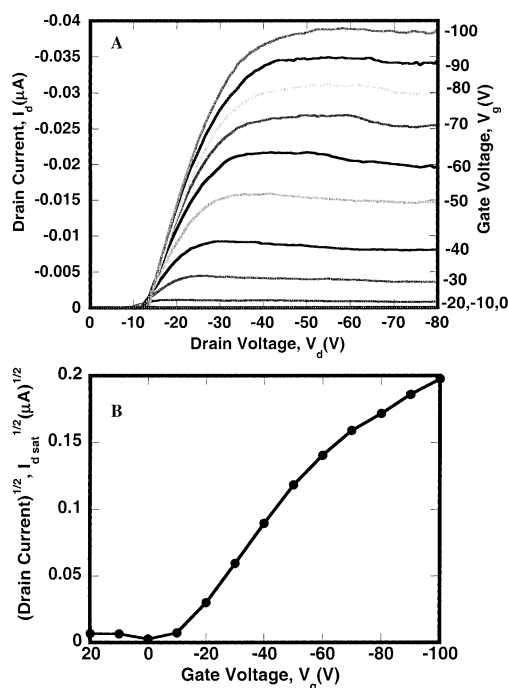**Figure 6.** Output characteristics (A) and $I_d^{1/2}$ versus V_g plot (B) of a PDT FET with $W = 400$ μm and $L = 20$ μm.

Figure 7 shows the FET output and transfer characteristics of a 50% blend. The $I-V$ curves show typical FET characteristics, namely, the I_d saturates at high V_d and is strongly modulated by the gate voltage.[1,20] The field-effect mobility calculated using eq 1 and a plot of $I_d^{1/2}$ vs V_g (Figure 7B) was 0.003 cm^2/(V s) with an I_{on}/I_{off} ratio of 3×10^2. This field-effect mobility of holes in the 50 wt % blend is about a factor of 6.7 lower than the mobility in PHT homopolymer and about a factor of 38 larger than that of PDT homopolymer. The output characteristics of the 10% blend showed good drain current saturation; the field-effect mobility calculated for the saturation region was 0.004 cm^2/(V s) with an I_{on}/I_{off} ratio of 10^3. The

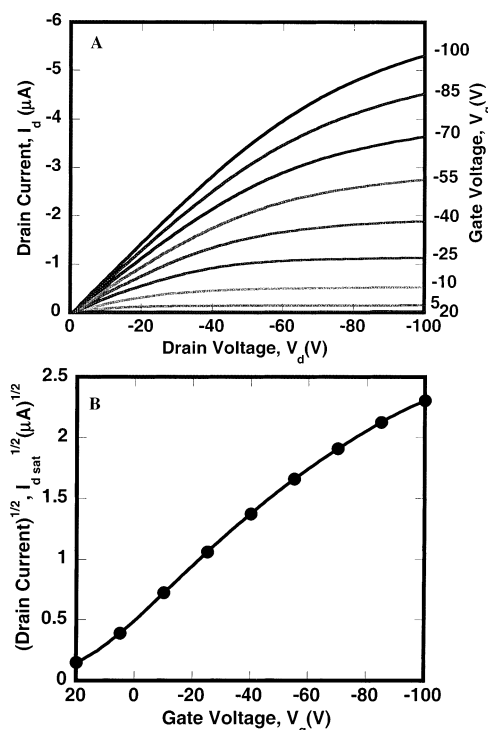**Figure 7.** Output characteristics (A) and $I_d^{1/2}$ versus V_g plot (B) of an FET with $W = 1500$ μm and $L = 50$ μm made from 50% PDT blend.
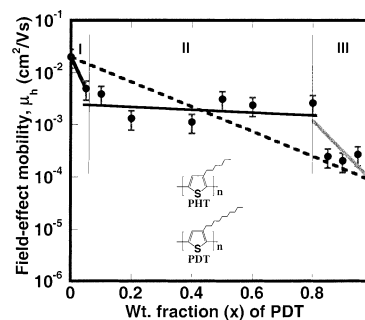

Figure 8. Blend composition dependence of the field-effect mobility of holes in PHT/PDT blends. The dashed line represents an exponential fit.

TABLE 1: Field-Effect Transistor Properties of PHT/PDT Blend System

material	μ_h, (cm^2/(V s))	I_{on}/I_{off}
PHT (0 wt % PDT)	2×10^{-2}	1×10^2
5 wt % PDT	5×10^{-3}	1×10^2
10 wt % PDT	4×10^{-3}	1×10^3
20 wt % PDT	1×10^{-3}	2×10^3
40 wt % PDT	1×10^{-3}	1×10^1
50 wt % PDT	3×10^{-3}	3×10^2
60 wt % PDT	2×10^{-3}	1×10^2
80 wt % PDT	3×10^{-3}	2×10^1
85 wt % PDT	2×10^{-4}	1×10^3
90 wt % PDT	2×10^{-4}	1×10^4
95 wt % PDT	3×10^{-4}	1×10^4
PDT (100 wt % PDT)	8×10^{-5}	3×10^3

80% blend similarly showed a field-effect mobility of 0.003 cm^2/(V s) for the saturation region. The field-effect mobility of holes and the on/off current ratio measured for all the blends and homoploymers are collected together in Table 1.

Blend Composition Dependence of Hole Mobility. The composition-dependent field-effect mobility of holes ($\mu_h(x)$) in the PHT/PDT blend system is shown in Figure 8, where x is the weight fraction of the lower-mobility component (PDT).

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The expected composition dependence of hole mobility in a disordered organic solid where charge transport is described by energetic disorder controlled hopping is a power law or an exponetial function of the form $\mu_h(x) = \mu_o \exp(-ax)$.[22] Representation of such a dependence on a semilog plot, as in Figure 8, would predict that the blend data points fall on a linear connection between the homopolymer data points. However, an exponential fit (the dashed line in Figure 8) did not describe the data well. The large departure of the mobility from exponential function in the 0.2 to 0.8 PDT weight fraction range gives an unusual "stair step" appearance to the $\mu_h(x)$ data. The entire blend composition can be divided into three regions of distinct features of the hole mobility. In region I, the field-effect mobility is decreased by a factor of 5−7 compared to the PHT homopolymer. Because PHT and PDT are identical in all respects (e.g., the HOMO and LUMO levels are identical, the π-stacking distance is the same in both) except the interlayer d spacing of their layered crystalline structures,[15a,20] the large decrease in mobility upon addition of only 5% PDT should be rationalized in terms of structural disorder involving this specific factor.

The field-effect mobility of holes in the 5−80 wt. % composition range, region II, is found to be relatively high and constant at about 2×10^{-3} cm²/(V s) (Figure 8). Achievement of such a high mobility in a series of many blends is remarkable because there are very few conjugated polymer semiconductors with carrier mobility as high as 10^{-3} cm²/(V s).[1−9] On the basis of our AFM studies of these PHT/PDT blends and prior X-ray diffraction studies of related binary blends of regioregular poly-(3-alkylthiophene)s,[15] these blends in the 5−80% composition range are homogeneous single-phase crystalline materials. The interlayer d spacing of these blends must be the same and intermediate between those of PHT and PDT as found in blends of PHT and poly(3-octylthiophene),[15a] leading to the observed high mobility in region II. These results clearly demonstrate that blends of conjugated polymers are alloy semiconductors capable of having high charge carrier mobility. This means that blending should also be considered a viable supramolecular engineering approach to enhancing or controlling the carrier mobility in conjugated polymer semiconductors.

The hole mobility in blends with greater than 80% PDT (region III) was found to decrease exponentially with composition. Nevertheless, the hole mobility of blends in this composition region is still larger than that of pure PDT. If one accepts the current explanation that the reason the hole mobility of PDT is substantially smaller than that of PHT is because of the larger interlayer d spacing of its layered crystalline structure,[1,5,8] then blends in this composition region (80% < x < 100%) must be such that the presence of varying amounts of PHT progressively reduces the interlayer spacing and thus resulting in the observed increases in carrier mobility. The available X-ray diffraction data for PHT/poly(3-octylthiophene) blend system indeed show such a composition-dependent interlayer d spacing.[15a] Although the composition dependence of the field-effect hole mobility in the region can be described by an exponential function, it is not clear that the energetic disorder controlled hopping model[22] is applicable even in this narrow composition region.

The observed "stair step" composition dependence of the field-effect mobility of holes in the present blends of crystalline regioregular poly(3-alkylthiophene)s and particularly the different regions of hole mobility has been largely rationalized in terms of variation in the crystalline structure of the blends. However, a similar "stair-step" blend composition dependence of the field-effect mobility was observed in n-channel thin film transistors based on amorphous binary blends where *electrons* were the charge carriers.[9b] These earlier observations and the present results suggest that the "stair step" composition dependence of the field-effect mobility of charge carriers may be a general phenomenon and characteristic feature in the charge transport in blends of conjugated polymer semiconductors. Investigation of other blend systems should be done to confirm the generality of this composition dependence of carrier mobility in blends of conjugated polymers. It is known that the morphology of the material in the channel region near the contact electrode may limit the device performance.[1c] To address this issue, further studies of top contact geometry will be pursued in the future.

Conclusions

Charge carrier transport in a series of binary blends of regioregular poly(3-alkylthiophene)s was investigated by fabricating and characterizing thin film transistors based on the alloy semiconductors. The measured field-effect mobility of holes in regioregular poly(3-hexylthiophene) (PHT) homopolymer, 0.02 cm²/(V s), was comparable with the previously reported highest values.[5−7] The field-effect hole mobility in the PHT/PDT blends had a "stair step" dependence on blend composition, resulting in three distinct regions of $\mu_h(x)$. The hole mobility in the 5−80 wt % blends was relatively high and constant at about 2×10^{-3} cm²/(V s) and was explained in terms of single-phase crystalline materials with a composition-independent interlayer d spacing. AFM results confirmed the homogeneous single-phase nature of the blends. In blends containing greater than 80 wt % PDT the hole mobility decreased exponentially with composition.

The present results demonstrate that blends of conjugated polymer semiconductors provide an approach to investigating the morphology−charge transport relationships of interest to not only thin film transistors but also other areas of polymer electronics such as light emitting diodes and photovoltaic cells.

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