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Controlling organization of conjugated polymer films from binary solvent mixtures for high performance organic field-effect transistors

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ABSTRACT

We investigate the effect of a binary solvent blend as a solvent for poly{[*N*,*N'*-bis(2-octyldodecyl)-1,4,5,8-naphthalenediimide-2,6-diyl]-alt-5,5'-(2,2'-bithiophene)} P(NDI2OD-T2) on the characteristics of n-channel organic field-effect transistors (OFETs). To make the binary solvent blend, the low-boiling-point non-solvent propylene glycol methyl ether acetate (PGMEA, b.p ~146 °C) is added to the high-boiling-point good solvent 1,2-dichlorobenzene (O-DCB, b.p ~180 °C) at various mixing ratio from 0 to 40 v%. UV–vis spectra of P(NDI2OD-T2) solution dissolved in the binary solvent clearly show the formation of polymer aggregates through a gradual red shift of the intramolecular charge transfer band with the addition of high concentrations of non-solvent PGMEA. Higher edge-on oriented crystallinity is observed for P(NDI2OD-T2) films spin-coated from the binary solvent with 5–10 v% PGMEA by out-of-order x-ray diffraction. P(NDI2OD-T2) films are applied as the active layer in top-gate/bottom-contact OFETs. Improved n-type field-effect mobility of the P(NDI2OD-T2) semiconducting layer up to 0.59 cm²/Vs was achieved for on-center spin coated films compared to 1.03 cm²/Vs for off-center (parallel alignment) spin-coated films respectively employing the binary solvent with 10 v% PGMEA.

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1. Introduction

Representing an enthralling domain in material science, the field of research pertaining to organic π -conjugated molecules has led to tremendous progress in the comprehension of intriguing physicalchemical processes for the development of innovative optoelectronic technologies complementary to inorganic counterparts [1–3]. Organic π -conjugated molecules have largely been adopted to develop flexible organic field-effect transistors (OFETs) thanks to their intrinsic soft matter properties [4,5]. Charge transport in deposited semiconducting films, which is known to have a significant dependence on organization of π -conjugated polymers, is critical to the performance of semiconducting polymer-based OFETs [6,7]. Various processing techniques such as dielectric surface treatments [8], optimization of film deposition methods [9], use of high-boiling-point solvents [10], solution aging [11], and postdeposition techniques such as solvent vapor and thermal annealing [12] have been explored to effect a change in the supramolecular assembly of π -conjugated polymers with a resulting positive impact on the charge-carrier transport properties of polymer OFETs.

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The use of binary solvent blends consisting of a non-solvent and a good solvent have recently been demonstrated to improve molecular ordering for facilitating charge transport in polymer semiconductor film without any additional post-treatment process [13–15]. In such binary-blend strategies, non-solvents with higher boiling points than those of the good solvents have been commonly used because of their potential to form more-ordered aggregates of π -conjugated polymers during solvent evaporation. In such systems, the less-volatile non-solvent resides within the evolving film for a longer period of time and therefore promotes supramolecular aggregation. Chang et al., observed no significant difference in charge transport between the P3HT film from a single good solvent and the P3HT film from binary solvents that included a low-boiling-point non-solvent [12]. However, the use of lowboiling-point non-solvents for binary solvent blends has rarely been investigated to improve ordering of π -conjugated polymers for high performance OFETs.

In this report, we investigate the effect of a binary solvent consisting of a low-boiling-point non-solvent and a high-boiling-point good solvent to produce poly{[N,N'-bis(2-octyldodecyl)-1,4,5,8-naphthalenediimide-2,6-diyl]-alt-5,5'-(2,2'-bithiophene)} P(NDI2OD-T2) films as the active layer in n-channel OFETs. In this study, we selected propylene glycol methyl ether acetate (PGMEA) (b.p. ~146 °C) as the non-solvent and 1,2-dichlorobenzene (O-DCB)

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(b.p. ~180 °C)] as the good solvent using the Hansen solubility parameter program (HSP). HSP analysis revealed an enhanced solvent-solvent and solvent blend-polymer interaction for this pair of solvents due to their similar interacting solubility parameters upon addition of up to 20 v% of the non-solvent. Marked effects on polymer crystallinity and morphological change of the spin-coated polymer film, evidenced by the formation polymer aggregates in both solution and film state, were observed upon varying the amount of PGMEA added in the blend solvent. P(NDI2OD-T2) films, for top-gate/bottom-contact (TG/BC) geometry OFET devices, fabricated from the binary solvent with 10 v% PGMEA by aligning the polymer film using an off-center (parallel alignment) spin-coating method and by a conventional on-center spin coating method showed an n-type field-effect mobility (μ) of up to ~ 1.03 cm²/Vs and ~0.59 cm²/Vs, respectively.

2. Experimental

2.1. Hansen solubility parameter analysis

5 mg of P(NDI2OD-T2) was dissolved in 1 ml of O-DCB/PGMEA solvent blends in the ratios 100:0, 95:5, 90:10, 80:20,70:30, 60:40, 50:50, 40:60, 30:70, 20:80, 10:90, and 0:100 v% according to the defined procedure for solubility parameter estimation, and heated overnight at 80^oC. Subsequently, the extent of solubility of the polymer and the solubility parameters were determined from these solutions via visual examination. Solvents were categorized as poor if they were unable to dissolve the polymer and good if they were able to dissolve the polymer at the stated temperature. Employing the binary solvent grid method in the Hansen solubility parameter (HSP) analysis, using the software Hansen Solubility Parameters in practice (fifth edition), a poor solvent was ascribed a value of "0" and good solvent was ascribed a value of "1". A sphere in Hansen space was created by the HSP software based on the coordinates of the good solvents, while excluding those of the poor solvents. The center of the sphere represented the HSP of the polymer P(NDI2OD-T2) with fitting accuracy of "1", obtained indicating the best fit.

2.2. OFET fabrication

The top-gate/bottom-contact device architecture was used in the OFETs fabrication. A standard photolithography technique was employed in patterning (Au/Ni = 13/3 nm) thick source and drain electrodes on a Corning Eagle XG glass substrates. The substrates were subsequently cleaned in an ultrasonic bath with solutions of acetone, isopropanol, and distilled water respectively for 10 minutes successively. The semiconducting polymer P(NDI2OD-T2) (ActivInk N2200; Polyera Inc.) was purchased from Polyera Corporation and 5 mg/ml of polymer solutions were prepared using O-DCB/PGMEA solvent blends with the ratios 100:0, 95:5, 90:10, 80:20 and 60:40 vol%. Using on-center and off-center spin coating methods, the resulting solutions were spin coated at 2000 rpm for 60s in an N₂-purged glove box. The films obtained were thermally annealed at 150 °C for 30 minutes and allowed to cool down slowly in a N₂-purged glove box. An 80 mg/ml solution of Poly(methyl methacrylate) (PMMA, Sigma-Aldrich, Mn: 120 kDa) in *n*-butyl acetate was spin-coated on the P(NDI2OD-T2) layer at 2000 rpm for 60 s and the resulting PMMA film was baked at 80 °C for 2 hours to form the gate dielectric layer. 50 nm aluminum (Al) as the gate electrode was thermally evaporated using a metal shadow mask under moderate vacuum conditions at a base pressure below 5×10^6 Torr to complete the device fabrication.

2.3. Characterization

P(NDI2OD-T2) OFET's electrical characteristics were measured using a Keithley 4200-SCS semiconductor parameter analyzer connected to an N₂-filled glove box probe station. The mobility (μ) and the threshold voltage (V_T) of the devices were determined from the saturation region using the gradual channel approximation method. The UV–Vis spectra of both solution and film states of P(NDI2OD-T2) were obtained using a UV–Vis spectrophotometer (Agilent, CARY 5000). The thin film state was prepared by spin coating polymer solutions onto a pre-cleaned glass substrate following the same conditions for OFETs device fabrication. Solution absorption spectra were obtained by dissolving P(NDI2OD-T2) in the various solvent blends and measured at a concentration of 0.05 mg/ml. Atomic force microscopy (AFM) images were recorded using a Nano Scope Multimode system (Veeco, instruments, Bruker) in the non-contact mode, to characterize the surface morphological microstructures of P(NDI2OD-T2) thin films. The structural ordering and crystallinity of P(NDI2OD-T2) in solvent blends spin-coated on SiO₂/Si substrates was investigated using a Rigaku RINT 2000 X-ray diffractometer (XRD) with Cu Ka radiation.

3. Results and discussion

We employed P(NDI2OD-T2) (Fig. 1a), one of the most researched n-channel polymer having peculiar properties, excellent charge transport characteristics [16], and shows different degrees of pre-aggregation in solution depending on the type of solvent adapted [17]. O-DCB (Fig. 1b), a typical high-boiling-point good solvent (b.p. ~180 °C, vapor pressure ~180.7 Pa at 25 °C), known for its strong solubility, crystalline film forming ability due to its slow evaporation rate, ability to induce high degrees of preaggregation of the P(NDI2OD-T2) polymer in both solution and film state, and a relative energy difference (RED) of less than 1 determined by HSP analysis, was considered as a good solvent [17,18] in the binary blend. Propylene glycol methyl ether acetate (PGMEA) (b.p. ~145 °C, vapor pressure ~506.6 Pa at 25 °C), an excellent solvent used in the dissolution of polymer dielectrics, orthogonal to the P(NDI2OD-T2) polymer [19], and a relative energy difference RED more than 1 determined by HSP analysis, was adapted as the low-boiling-point non-solvent. By virtue of its superior injection characteristics, the typical staggered TG/BC device architecture (Fig. 1c), with glass substrate, Au/Ni source-drain contacts, poly(methyl methacrylate) (PMMA) gate dielectric, and aluminum gate contact was used in this study. To form the P(NDI2OD-T2) film, we applied both on-center and off-center spin coating methods (Fig. 1d). The off-center spin coating technique has been actively used to produce highly aligned polymer films through channels of OFETs for improving field effect mobility [7,20].

The solubility behavior of the system was analyzed by probing into the dynamics of solvent-solvent and solute-solvent interactions. It has been observed that miscibility and interaction between entities in a particular system are enhanced when components have similar cohesive energy densities (E/V) [21]. Hansen solubility parameters (HSPs), which describe the total solubility behavior (δ) of a system (Equation (1)) as a result of three different contributing factors, atomic dispersive interactions (δ_D), permanent dipole molecular interactions (δ_P), and molecular hydrogen and bonding interactions (δ_H), were adopted for the study [22].

$$\delta^2 = \delta_H^2 + \delta_P^2 + \delta_H^2 \tag{1}$$

Analysis with the Abbott and Hansen software HSPIP gave solubility properties in a three-dimensional co-ordinate system with the three Hansen parameters of both solvent blends and polymer in

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