

Well defined double layers via binary solvent mixtures for highly efficient inverted all-polymer solar cells

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ABSTRACT

All-polymer solar cells have consistently attracted much attention to the planar heterojunction structures. However, key issues limiting the efficiency of bilayer all-polymer solar cells (BAPSCs) are their poor charge carrier transport and the strong possibility of recombination because of poorly organized double layer structures. Here we suggest well defined active systems via solvent engineering techniques for highly efficient BAPSCs. Thus, we systematically processed the single/co-solvents in each or both planar heterofilms, and investigated their morphological effects and device performances by charge carrier kinetics. Notably, more ordered molecular packing with stronger face-on stacking resulted from the co-solvent- treated active films, as revealed by atomic force microscopy and detailed two-dimensional grazing incidence wide-angle X-ray scattering measurements. Furthermore, these well-crystallized double layers enabled more efficient charge carrier movement, resulting in higher device efficiency. Our novel process offers an effective and facile method for precise control of the active layer morphology and a route to more efficient BAPSCs.

1. Introduction

All-polymer solar cells (APSCs) composed of polymer donors and polymer acceptors have been studied extensively as polymer:fullerene solar cell counterparts because of their superior mechanical strength, high flexibility, and thermal/photochemical stability [1–3]. Furthermore, all-polymer blends are readily processed in solution, are often inexpensive, and have good chemical and morphological stabilities resulting from entanglement of the polymers [4,5]. State-of-the-art APSCs are based on randomly mixed networks of donors and acceptors and have so-called bulk heterojunction (BHJ) architecture [6–8]. However, intrinsic polymer properties can lead to non-homogeneous blends, which can detrimentally affect the BHJ structure in APSCs. Hence, new materials [9,10] and/or additional morphological treatments [11–13] (e.g., additive processing) have been explored.

In contrast to BHJ models, APSCs with bilayer geometries can form clearer films having well-organized polymer molecular structures in separate active layers [14]. Control of the morphology can thereby be achieved at the nanostructural level, and well-oriented molecular domains can be maximized. Nevertheless, the efficiencies of bilayer APSCs (BAPSCs) have lagged behind those of BHJ-based APSCs, with the highest efficiency reaching only ca. 2% [15,16].

One strategy to boost the power conversion efficiency (PCE) of

BAPSCs toward those of BHJ-based APSCs is to control the nanostructure morphology of each polymer layer to facilitate better efficient charge transport and less charge recombination. In particular, well-oriented molecular packing such as face-on stacking can lead to much more efficient directional carrier movement, resulting in enhanced solar cell performance [17–20]. Motivated by these concepts, we undertook a detailed study of donor/acceptor double layer morphological control using solvent processing techniques. Our strategy was to optimize the nanostructural morphology with relatively well-ordered horizontally crystallites within large crystalline domains for individual active layers using different solvents, thereby enabling the layers to function as independent cells. We systematically investigated their crystalline structures using atomic force microscopy (AFM) and two-dimensional grazing incidence wide-angle X-ray scattering (2D-GIWAXS), and found that the co-solvents indeed enabled the formation of stronger face-on packing domains having more oriented molecular structures, which resulted in higher device performances as measured by higher photocurrents. We achieved a high PCE value of 2.94% under the simulated 1-sun AM 1.5G illumination condition, which we believe is the highest efficiency of a BAPSC reported to date. Our solvent engineering approach is a facile process that offers excellent molecular structure control and a route toward more efficient BAPSCs.

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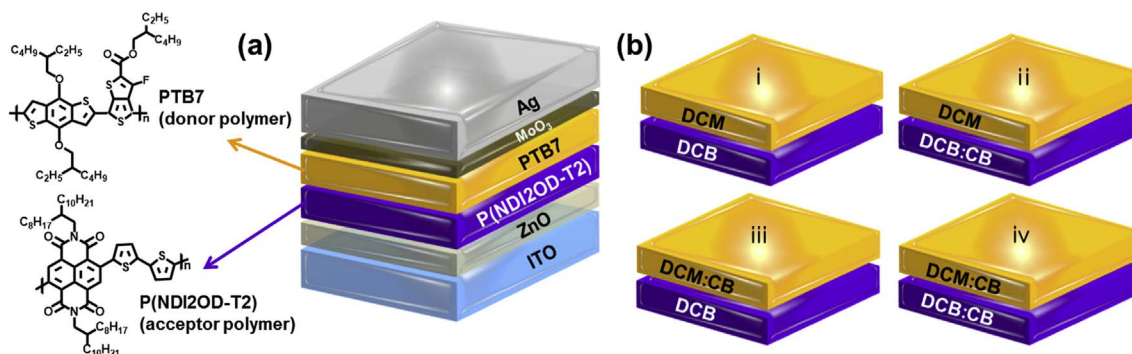


Fig. 1. (a) The chemical structures of PTB7 (donor polymer) and P(NDI2OD-T2) (acceptor polymer) in an inverted bilayer device and (b) four bilayer active systems made using the various processing solvents described in this work.

2. Results and discussion

2.1. Device architecture and model design

Poly{[4,8-bis[(2-ethylhexyl)oxyl]benzo[1,2-b:4,5-b']dithiophene-2,6-diyl][3-fluoro-2-[(2-ethylhexyl)carbonyl]thieno[3,4-b]thiophene-diyl]} (PTB7) was selected as the donor material because it can provide high hole mobility, a face-on stacking structure, and good device performance when blended with PC₇₁BM in general solar cell systems [21,22]. Poly{[N,N-bis(2-octyldodecyl)-naphthalene-1,4,5,8-bis(dicarboximide)-2,6-diyl]alt-5,5,9-(2,29-bithiophene)} [P(NDI2OD-T2)] was selected as the electron acceptor material because it can provide good electron movement along face-on packing structures [23,24]. These two active materials were used to fabricate inverted solar cells having the indium tin oxide (ITO)/ZnO/P(NDI2OD-T2)/PTB7/MoO₃/Ag architecture (Fig. 1a). The effect of different co-solvent systems on P(NDI2OD-T2)/PTB7 photovoltaic cells was investigated through the design of four different heterolayers: (i) dichlorobenzene (DCB)-processed P(NDI2OD-T2)/dichloromethane (DCM)-processed PTB7, (ii) DCB:chlorobenzene (CB)-processed P(NDI2OD-T2)/DCM-processed PTB7, (iii) DCB-processed P(NDI2OD-T2)/DCM:CB (2:1)-processed PTB7, and (iv) DCB:CB (2:1)-processed P(NDI2OD-T2)/DCM:CB-processed PTB7 (Fig. 1b). We found that such solvent engineering of the active P(NDI2OD-T2)/PTB7 layers greatly affected the performance of bilayer solar cells made with them.

2.2. Creating high performance bilayer all PSCs with the co-solvent approach

The photocurrent vs. voltage (J - V) behavior of four different double layer diodes was examined to explore the solvent processing effect. Fig. 2a shows the performances of the inverted solar cells having well-defined P(NDI2OD-T2)/PTB7 bilayers processed with and without a co-

solvent; their corresponding photovoltaic parameters are listed in Table 1. For case (i) where each film in the bilayer was processed in single, but different, solvents (i.e., DCB/DCM), the J - V curve showed that this cell had a relatively low device efficiency of 1.47% with the following parameters: open-circuit voltage (V_{oc}) = 0.72 V, short-circuit current (J_{sc}) = 5.15 mA cm⁻², and fill factor (FF) = 42.3%. Notably, when the P(NDI2OD-T2) [case (ii); DCB:CB/DCM] or PTB7 [case (iii); DCB/DCM:CB] layer was treated with co-solvents, the performance of the bilayer devices exhibited improved performance, i.e., PCE = 2.05%, V_{oc} = 0.72 V, J_{sc} = 6.28 mA cm⁻², and FF = 45.4% for case (ii) devices and PCE = 2.48%, V_{oc} = 0.73 V, J_{sc} = 6.43 mA cm⁻², and FF = 52.7% for case (iii) cells. In comparison with cases (ii) and (iii), the solar cells made with the PTB7 layer processed with the co-solvent were better performing, which indicated a positive effect of the co-solvent on the PTB7 of the P(NDI2OD-T2)/PTB7 system. Surprisingly, the best PCE of 3.05% was measured for case (iv) of the inverted P(NDI2OD-T2)/PTB7 double layer; it also had the highest J_{sc} of 7.61 mA cm⁻² and FF of 54.1%. This was the largest PCE enhancement achieved via co-solvent treatments for both layers (DCB:CB/DCM:CB). Furthermore, this efficiency and photocurrent are among the highest reported for BAPSCs. In addition, in comparison with BHJ-based P(NDI2OD-T2)/PTB7 solar cells (near 2%) [25,26], our bilayer system showed higher performances. These results suggest that the phase morphology had altered to more ordered structures, and that the carriers could more easily travel to the electrodes [27,28].

The external quantum efficiency (EQE) curves of our APSCs based on bilayer structures are shown in Fig. 2b. Among the four different solvent-processed devices, the DCB:CB/DCM:CB co-solvent-treated P(NDI2OD-T2)/PTB7 double layer case displayed the highest response when illuminated. The EQE responses followed the trend in J_{sc} values of the photovoltaic cells [29].

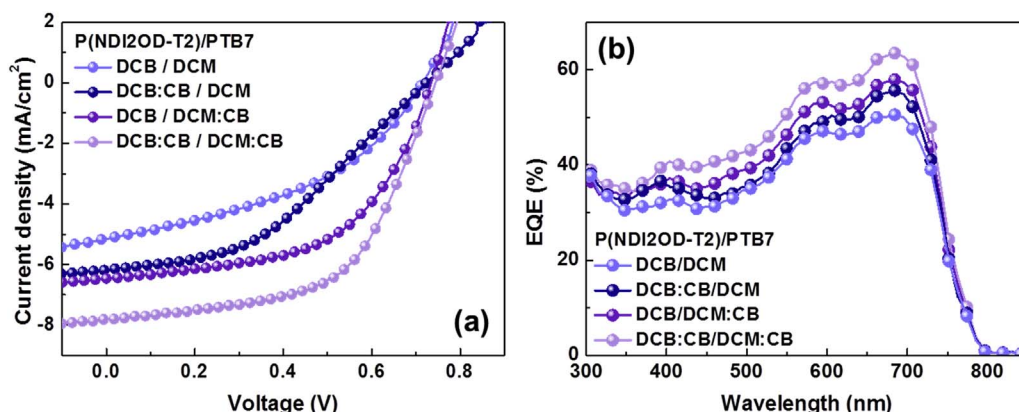


Fig. 2. (a) Illuminated J - V curves of different P(NDI2OD-T2)/PTB7 inverted photovoltaic systems and (b) their corresponding external quantum efficiency (EQE) spectra.

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