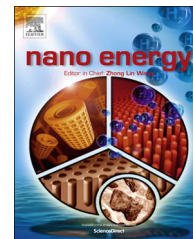




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RAPID COMMUNICATION

Nano-pathways: Bridging the divide between water-processable nanoparticulate and bulk heterojunction organic photovoltaics



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Abbreviations: BHJ, bulk heterojunction; DCB, dichlorobenzene; EQE, external quantum efficiency; FF, fill factor; ITO, indium tin oxide; J_{sc} , short circuit current density; M_n , number average molecular weight; M_w , weight average molecular weight; NP-OPV, nanoparticle organic photovoltaic; P3HT, poly(3-hexylthiophene); PC₆₁BM, (phenyl C₆₁ butyric acid methyl ester); PC₇₁BM, (phenyl C₇₁ butyric acid methyl ester); PCE, power conversion efficiency; PDI, polydispersity; PEDOT:PSS, poly(3,4-ethylenedioxythiophene) polystyrene sulphonate; R2R, roll-to-roll; RPV, Resistance dependent PhotoVoltage; SDS, sodium dodecyl sulphate; SEC, size exclusion chromatography; SEM, scanning electron microscopy; Si₃N₄, silicon nitride; STXM, scanning transmission X-ray microscopy; TEM, transmission electron microscopy; T_g, glass transition temperature; TQ1, poly[2,3-bis-(3-octyloxyphenyl)quinoxaline-5,8-diyl-alt-thiophene-2,5-diyl]; V_{oc}, open circuit voltage

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KEYWORDS

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Organic photovoltaic;
Blend morphology;
Glass transition temperature;
Scanning transmission X-ray microscopy

Abstract

Here we report the application of a conjugated copolymer based on thiophene and quinoxaline units, namely poly[2,3-bis-(3-octyloxyphenyl)quinoxaline-5,8-diyl-alt-thiophene-2,5-diyl] (TQ1), to nanoparticle organic photovoltaics (NP-OPVs). TQ1 exhibits more desirable material properties for NP-OPV fabrication and operation, particularly a high glass transition temperature (T_g) and amorphous nature, compared to the commonly applied semicrystalline polymer poly(3-hexylthiophene) (P3HT). This study reports the optimisation of TQ1:PC₇₁BM (phenyl C₇₁ butyric acid methyl ester) NP-OPV device performance by the application of mild thermal annealing treatments in the range of the T_g (sub- T_g and post- T_g), both in the active layer drying stage and post-cathode deposition annealing stage of device fabrication, and an in-depth study of the effect of these treatments on nanoparticle film morphology. In addition, we report a type of morphological evolution in nanoparticle films for OPV active layers that has not previously been observed, that of PC₇₁BM nano-pathway formation between dispersed PC₇₁BM-rich nanoparticle cores, which have the benefit of making the bulk film more conducive to charge percolation and extraction.

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Introduction

Organic photovoltaics (OPVs) offer an attractive, renewable energy solution to the demands of the planet's growing population in that they can be manufactured by high throughput methods reducing the fabrication costs, and are light-weight and flexible increasing their range of potential applications. In the last decade impressive results have been achieved for bulk heterojunction (BHJ) OPVs, such as power conversion efficiencies (PCEs) exceeding the 10% benchmark [1] and projected lifetimes of OPV on rigid substrates of over 7 years [2].

As the roll-to-roll (R2R) printing process is *the* envisioned technique for high-throughput manufacturing of OPVs, so-called solar inks need to be produced which, however, are often based on harmful chlorinated solvents such as chloroform, chlorobenzene or dichlorobenzene. More recently, the OPV community has switched to less harmful solvents like toluene, *o*-xylene, mesitylene or mixtures thereof [3-6], but much more ideal would be inks based on green solvents such as ethanol or the absolute champion solvent, water.

The relatively recent introduction of nanoparticle organic photovoltaics (NP-OPVs) offers several advantages; the first and foremost being the manufacturing of OPVs from easily produced water-based solar inks [7]. Furthermore, an additional level of morphological control of the photoactive layer can be achieved [8-10], an appealing feature due to the importance of morphology to OPV performance [11-14] which involves a delicate balance between (1) mixing on a sufficiently small scale to provide a large surface area of contact between donor and acceptor materials for efficient exciton dissociation, and (2) mixing to be such that a network of connected donor and acceptor nano-domains exists to provide charge percolation pathways for holes and electrons to the anode and cathode, respectively [15]. In addition, the production of NP-OPV solar inks does not require any chemical modification of conjugated polymer backbones [16,17], which simplifies material synthesis. Even though NP-OPVs are yet to reach the efficiencies of their

BHJ counterparts, the rapid improvement of device PCEs from 0.55% [7] to 4.1% [18] in a mere three years, forecasts a fast catchup to BHJ OPV performance in the near future.

The production of NP-OPVs requires thermal treatment after deposition of the solar ink to remove water and to sinter the nanoparticles together to form a continuous interconnected film [8,9,18,19]. Previous work has shown that the glass transition temperature (T_g) of the semiconducting materials plays a major role in determining the optimal annealing conditions for tuning the final morphology of the photoactive layer blend film [20-23]. If the material T_g is too high, this can lead to difficulty in annealing the material films without degrading other components of the OPV devices [24]. But even more importantly, if the T_g is too low (below OPV operating temperatures which can reach up to 85 °C [25]) severe phase segregation can occur and a reduction in PCE results. The T_g also needs to be sufficiently high to allow mild drying treatments to remove residual casting/printing solvent without affecting film morphology [9]. The crystallinity of the polymer material is a second material property that is relevant to film morphology; blend films containing a semicrystalline polymer, such as poly(3-hexylthiophene) (P3HT), can undergo excessive phase separation rapidly during annealing treatments [26]. Additionally, a predominantly core-shell structure is retained with NP-OPVs based on P3HT [8,9,27], and depending on the purity of the domains, this structure is not always ideal for efficient charge transport. To sinter NP-OPVs, the blend needs to exhibit some viscous flow during thermal treatment. This property is more easily achievable in amorphous materials as their viscosity drops more rapidly over T_g while crystalline polymers such as P3HT incorporate physical crosslinks (crystallites) that restrict the required viscous flow.

TQ1 [28], possessing a high T_g (~100 °C) [29] and non-crystalline nature [24,30,31], has herein proven to be an interesting candidate for NP-OPV device fabrication. TQ1, in a blend system with PC₇₁BM, has facilitated the development of nanoparticle film morphologies that promise to be

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