

Influence of crystallizable solvent on the morphology and performance of P3HT:PCBM bulk-heterojunction solar cells

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ABSTRACT

In this work, we present the effect of 1,3,5-trichlorobenzene (TCB) on the morphology and performance of a 1:1(w/w) rr-P3HT:PCBM polymer/fullerene bulk-heterojunction solar cell. The UV–vis spectra of the rr-P3HT:PCBM polymer/fullerene formed from a blend with various TCB concentrations (0–28 mg/ml) indicate an enhanced absorption within the 500–600 nm wavelength range when formed in the presence of the TCB. By increasing the concentration of TCB in the blend, the resultant spun-cast film morphology was improved due to grain size reduction, and the polymer film quality can be further improved via an annealing process. Moreover, the device efficiency is also improved due to a good quality thin film nanostructure. With the optimized TCB quantity and annealing temperature, the power conversion efficiency (PCE) achieved with the solar cell was up to 20% better than that attained by a similar device without the presence of TCB.

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1. Introduction

Organic based composite photovoltaic cells have attracted increasing attention as a renewable energy resource. Although inorganic semiconductors are the archetype for the fabrication of high efficiency solar cells, the cost of the manufacturing processes of conventional inorganic photovoltaics limits their application in many areas. The ability to process many organic conjugated materials in isolation may allow such devices to be made with a low fabrication cost and ease of manufacture, and as a light weight product with a large area that is compatible with flexible substrates [1–3]. The low cost processes also include various common printing technologies such as ink-jet or doctor blade screen-printing. Therefore, a new range of electronic devices can be attached to unexpected elements, such as on paper sheets or garment products, due to benefit of plastic substrates and innovative conductive inks [4–6]. Unfortunately, two major disadvantages of organic solar cells are the low overall power conversion efficiency (PCE), particularly true for large area devices and their stability or lifetime compared with conventional inorganic solar cells, such as silicon solar cells. However, the PCE and lifespan of organic photovoltaic cells (OPVs) have been dramatically improved in the past decade [7–9]. One of the most extensively investigated and promising material combination in

polymer solar cells are that of regioregular poly(3-hexylthiophene) (rr-P3HT) with a fullerene derivative ([6,6]-phenyl-C61 butyric acid methyl ester, PCBM) as the donor and acceptor materials, respectively, in a bulk-heterojunction [10,11]. The bulk-heterojunction PVCs made from a blend of rr-P3HT and PCBM have recently been shown to accomplish a PCE up to 3–5% and more than 6% for tandem cells [2,12–14]. In order to achieve the high photovoltaic performance, the band gap energy, HOMO/LUMO levels and carrier mobilities of the heterojunction materials need to be optimized simultaneously. The types of solvent or annealing process in the production of the active materials can also enhance the performance. Therefore, the different device geometries and interface morphologies need to be evaluated for the purpose of trapping more light, dissociating excitons more efficiently, increasing the exciton diffusion length and transporting charges with fewer barriers.

To obtain higher photocurrents, the typical length scale of phase separation between the donor and the acceptor phases should be in the order of the exciton diffusion length, which is around 10 nm for organic semiconductors [15]. The self-alignment ability of rr-P3HT facilitates the formation of thin films with well-ordered lamellae of co-facially stacked planarized main chains. This assembly ensures a fast charge transport in the layer of the ordered backbones, result in the achievement of a high charge carrier mobility. To obtain this condition, apart from the annealing process, various solvent selections have been used, such as chlorobenzene, 1,2-dichlorobenzene, 1,2,4 trichlorobenzene, chloroform and toluene. Among the choice of solvents,

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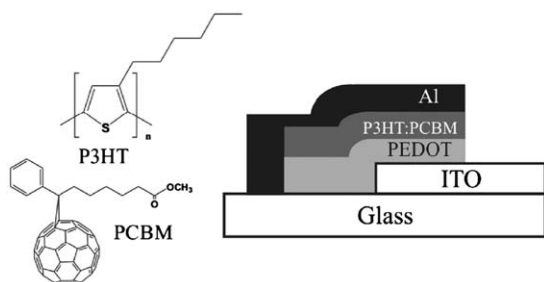


Fig. 1. Schematic of the device structure of a polymer bulk-heterojunction photovoltaic cell, and the chemical structures of the two main components.

chlorobenzene and its derivatives can produce a good device performance when compared with toluene [16–18]. Hereby, most additives in the liquid form with a high boiling point as co-solvent are applied during the dissolving process to optimize the thin film morphology, such as a solvent mixing between chlorobenzene and chloroform or chlorobenzene and alkanedithiols, that can lead to a small grain boundary and resulting in a high photocurrent [16,19]. Brinkmann et al. proposed a new method to obtain thin films of oriented and nanostructured materials, which are of potential interest for the synthesis of active layers of solar cells, using the directional epitaxial solidification of rr-P3HT in crystalline solvent to yield highly oriented and crystalline P3HT thin films [20,21].

Here, we report our results for heterojunction organic solar cells based on a 1:1(w/w) blend of the regioregular polymer, rr-P3HT, to the fullerene, PCBM, where the active layers are deposited by spin-coating of solutions using chlorobenzene, with different concentrations of 1,3,5-trichlorobenzene (TCB; 0–28 mg/ml), as crystallizable aromatic solvents. We correlate these results with atomic force microscopy (AFM) images of the morphologies of the formed heterojunction materials. Fig. 1 schematically shows the typical structure of a polymer photovoltaic cell fabricated in this study, and the chemical structures of the used materials. Details of the device fabrication process and the characterization techniques are given in the experimental part.

2. Experimental details

2.1. Materials

All reactions were carried out under an argon atmosphere. Regioregular poly(3-hexylthiophene) was prepared by the Grignard metathesis procedure according to McCullough [22], which involves the polymerization of the Grignard reagent 2-bromo-5-bromomanesio-3-hexylthiophene. The average molecular weight (Mw) of P3HT was determined by GPC analysis against polystyrene standards and found to be 44,700 g/mol with a polydispersity index (PDI) of 1.53. PCBM was synthesized from the reaction of fullerene and methyl-4-benzoylbutyrate-*p*-tosyl-hydrazone, in close analogy to the method already described in the literature [23].

2.2. P3HT/PCBM ratio and concentration

The blend solution of bulk-heterojunction solar cells had a P3HT/PCBM ratio of 1.0:1.0 (wt/wt%); with a concentration of 10 mg/ml P3HT plus 10 mg/ml PCBM and TCB (Aldrich) at 0, 6, 12, 20 and 28 mg/ml in chlorobenzene.

2.3. Calibration and measurement

The optical absorption spectrum was investigated by means of UV–vis spectroscopy (Perkin Elmer, Lambda 650). The surface morphological features of the films were examined using a Seiko Instruments SPA-400 atomic force microscope (AFM) system simultaneously. The current density–voltage (J – V) characteristics of the bulk-heterojunction solar cells were measured under illumination at 100 mW/cm² using an AM 1.5 solar simulator under ambient conditions. The solar cells were illuminated through the side of the ITO-coated glass plate.

2.4. Device fabrication

Polymer solar cells were prepared according to the following procedure: The ITO-coated glass substrate (with a sheet resistivity of 15 Ω/sq, Semiconductor wafer, Inc) was first sequentially cleaned with detergent, methanol, acetone and isopropyl alcohol for 15 min, in an ultrasonic bath and subsequently dried with N₂. Conductive grade PEDOT:PSS (Aldrich; conductive grade) was spun-cast (2000 rpm) from an aqueous solution (after passing through a 0.45 μm filter). The substrate was dried for 60 min at 80 °C under reduced pressure and then spun-cast as the bulk-heterojunction layer. All P3HT and PCBM blend solutions were stirred for 24 h before spin-coating to ensure that the PCBM has completely dissolved in the P3HT. The chlorobenzene and TCB solution, used as a crystallizable solvent, was composed of 1% (w/w) P3HT and 1% (w/w) PCBM and was then spin-cast at 1500 rpm on top of the PEDOT:PSS layer. The active layer was annealed at 63, 110, 150 and 180 °C (TCB melting point=63 °C) for 30 min under a nitrogen atmosphere. Subsequently, TCB was removed by evaporation under reduced pressure. Finally, the samples were transferred into the vacuum chamber to deposit Al (~200 nm thick) on top of the spin-coated polymer film. The deposited Al electrode area defined the active area of the devices as 0.18 cm².

3. Results and discussion

The bulk-heterojunction approach (formed by blending donor type conjugated polymers with the acceptors like fullerenes) is attractive and offers significant progress in improving the PCE of these devices. One of the possible improvements in this type of solar cell is the implementation of a material that can absorb the red and near infrared part of the solar spectrum, where the maximal photon flux of the sun is located. Consequentially, this has been one of the fundamental strategies and main focus in improving the performance of organic solar cells. One of the advantages of polymer photovoltaic devices is the solution process ability of the active materials. However, the solvents used for the preparation of the active layer have a strong impact on its morphology, which in turn influences the performance of the solar cells [24]. The atomic force microscopy (AFM) topography images indicate the smoothed film surface of the P3HT:PCBM polymer/fullerene as the TCB concentration in the blend is increased, as shown in Fig. 2 for data obtained with an annealing temperature of 150 °C. The grain sizes are continuously reduced as the TCB concentration is increased up to 12 mg/ml, and thereafter the roughness increases as the TCB concentration is increased further. Therefore, only the roughness development can be observed as the TCB concentration is varied. This tendency can also be observed with all the other tested annealing temperature regimes, as summarized in Table 1.

From the data summarized in Table 1, the rms roughness is abruptly reduced as the TCB is added, particularly at the 63 °C (TCB

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