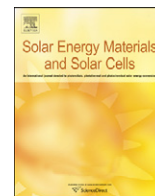




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Solar Energy Materials & Solar Cells

journal homepage: www.elsevier.com/locate/solmat

Interface modification of a highly air-stable polymer solar cell

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ARTICLE INFO

Article history:

Received 5 August 2011

Received in revised form

12 November 2011

Accepted 19 November 2011

Available online 7 December 2011

Keywords:

Polymer solar cells

Inverted structure

Stability

Interface modification

Solution-process

ABSTRACT

Stability is an essential issue in polymer solar cells for achieving commercially applicable devices. In this work, we report on a highly stable polymer solar cell, based on an inverted structure with the active layer sandwiched between a pair of metal-oxide buffer layers and an inherently air-stable conjugated polymer, a-PTPTBT. After interface modification, the unencapsulated device can retain power conversion efficiency without apparent degradation even after six months of storage in ambient conditions. This device's excellent stability is attributed to the air-stable and chemically stable conjugated polymer, as well as the environmentally robust metal oxides used as the interfacial layers. To further improve the device performance, cathode-side interface modification was also carried out. After the insertion of the TiO₂ nanorod interlayer between the ZnO thin film and the active layer, the device efficiency can be successfully increased from 3.3% to 4.1%. The future application of this materials system in inverted structures needs efforts devoted to morphology control and interface engineering to improve the device fill factor. The methodology applied here can serve as an effective strategy for fabricating highly air-stable devices with different materials systems.

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1. Introduction

Polymer photovoltaics (PV) have attracted considerable attention due to the potential of achieving flexible and light-weight devices over a large area with low fabrication costs [1–3]. To reach the goal of commercialization, three important aspects have to be considered. Efficiency, stability and the processing of the devices should be equally weighed and simultaneously optimized to reduce the unit energy costs of polymer photovoltaics [3–5]. Solution processing, which significantly simplifies the fabrication processes and lowers the costs, has been successfully applied to polymer solar cells by many research groups [6–15]. In this scenario, the active layer, the cathode or anode buffer layers [6–11], and even the electrodes [12–15] can be processed with solution. In addition to the fabrication process, most of the prevailing works are concerned mainly about the device power conversion efficiency (PCE) [16–19]. Little research has been devoted to the stability of devices. Typical devices employing poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS) as the anodic buffer layer and aluminum as the cathode

suffer from instability problems. Acidic PEDOT:PSS would corrode the indium–tin oxide (ITO) [20,21], whereas the low work function aluminum cathode is easily oxidized in air [22,23]. In addition, aluminum may diffuse into and react with the adjacent active layer [24,25]. All of these properties contribute to the degradation of devices, retarding the possibility of practical use. Finally, atmospheric oxygen and water may diffuse into the device through the pinholes or grains of the metallic back electrode and are regarded as another major source of degradation [4,26].

To circumvent the instability problems, several strategies can be employed to improve the device stability. The most intuitive and effective approach is to adopt encapsulation techniques, which require very stringent conditions if the device by itself does not have good stability [27]. Therefore, improving device stability before encapsulation is also very important. The inverted geometry for the devices is a good approach. Inverted devices, which employ high work function metals as the back electrode and electron selecting metal oxides for modifying ITO, have been reported to exhibit much higher stability than conventional ones [22,28–30]. The inverted devices can demonstrate sustainable PCEs even after long exposure to ambient conditions. For poly(3-hexylthiophene) (P3HT) and (6,6)-phenyl C61 butyric acid methyl ester (PCBM) based inverted cells, a 90% retention of PCE after 1000 h of storage in air without encapsulation has been reported [31].

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However, the devices may still undergo performance degradation in spite of the use of protective techniques or inverted structures. Atmospheric oxygen and water can still penetrate into the active layer across the protective layers, though at a much slower rate [4,32]. Therefore, the fundamental problem associated with the degradation lies in the instability of the conjugated polymer used in the active layer. Air-stable conjugated polymers have been reported in the application of organic thin-film transistors (OTFTs) to improve device stability [33–36]. To possess sufficient stability toward oxidative doping in ambient conditions, conjugated polymers should have a higher ionization potential, corresponding to a lower highest occupied molecular orbital (HOMO) level [37,38]. In addition to the enhanced stability, polymers with low-lying HOMO levels can generate higher open-circuit voltage (V_{oc}) when forming junctions with PCBM or (6,6)-phenyl C71 butyric acid methyl ester (PC₇₀BM). Herein we report on highly air-stable polymer photovoltaics based on an alternating copolymer consisting of a thiophene-phenylene-thiophene (TPT) electron-donating unit and a 2,1,3-benzothia-diazole (BT) electron-accepting moiety (a-PTPTBT) [39,40]. The molecular structure of a-PTPTBT is shown in Fig. 1. The combination of a-PTPTBT and the inverted geometry yields a highly air-stable

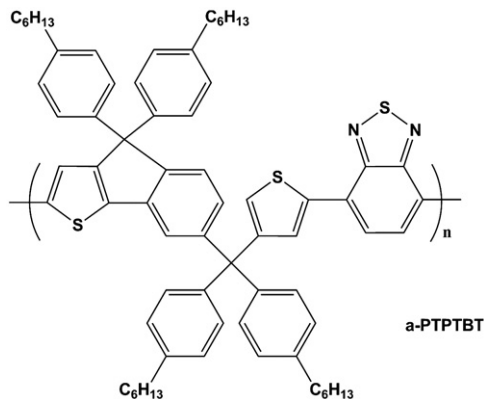


Fig. 1. The molecular structure of a-PTPTBT.

photovoltaic device that can retain its PCE without apparent degradation after long-term storage under ambient conditions. Further interface modification on the cathode side is also demonstrated and found to be effective in improving the device performance.

2. Experimental

Fig. 2a shows the schematic of the inverted device. The active layer (a-PTPTBT:PC₇₀BM) is sandwiched between and protected by a pair of metal-oxide buffer layers. The cathode buffer layer chosen here is a sol-gel derived zinc oxide (ZnO) thin film that displays reasonable electron mobility and selectivity even in the low-temperature deposition process. On the anode side, nickel(II) oxide nanoparticles are used, serving as the electron blocking layer and capable of modulating the current flow. The NiO anodic interlayer has been reported to function effectively as the hole transport layer (HTL) in either conventional or inverted devices [41–43]. However, most of the processes for depositing NiO need to be conducted in a vacuum. In contrast, the NiO nanoparticles used here can be solution-processed, so the process is simpler and less expensive. The energy level diagram of the functional layers and the active materials, as well as the electrodes, is shown in Fig. 2b.

All the devices were fabricated on pre-cleaned ITO coated glass substrates (ITO-glass, 7 Ω /sq). A 0.5 M sol-gel solution containing equimolar zinc acetate and ethanolamine in 2-methoxyethanol was used to deposit ZnO thin films. The solution was spin-casted onto the ITO-glass and annealed at 200 °C for 1 h to derive a dense ZnO film. Subsequently, the devices were transferred into a nitrogen-filled glove box. The a-PTPTBT blended with PC₇₀BM was used as the active layer. A blend solution of a-PTPTBT and PC₇₀BM (1:2.5 w/w) dissolved in *o*-dichlorobenzene was then spin-casted on top of the ZnO layer and the device was moved into a Petri dish. After film drying and upon solvent annealing, the film showed a gray color. NiO nanopowder (Aldrich, < 50 nm, 99.8%) uniformly suspended in isopropyl alcohol (0.1 mg/ml) was spin-casted onto the active layer, and the device was then dried at room temperature for 10 min. Finally, the silver back electrode (~200 nm) was deposited

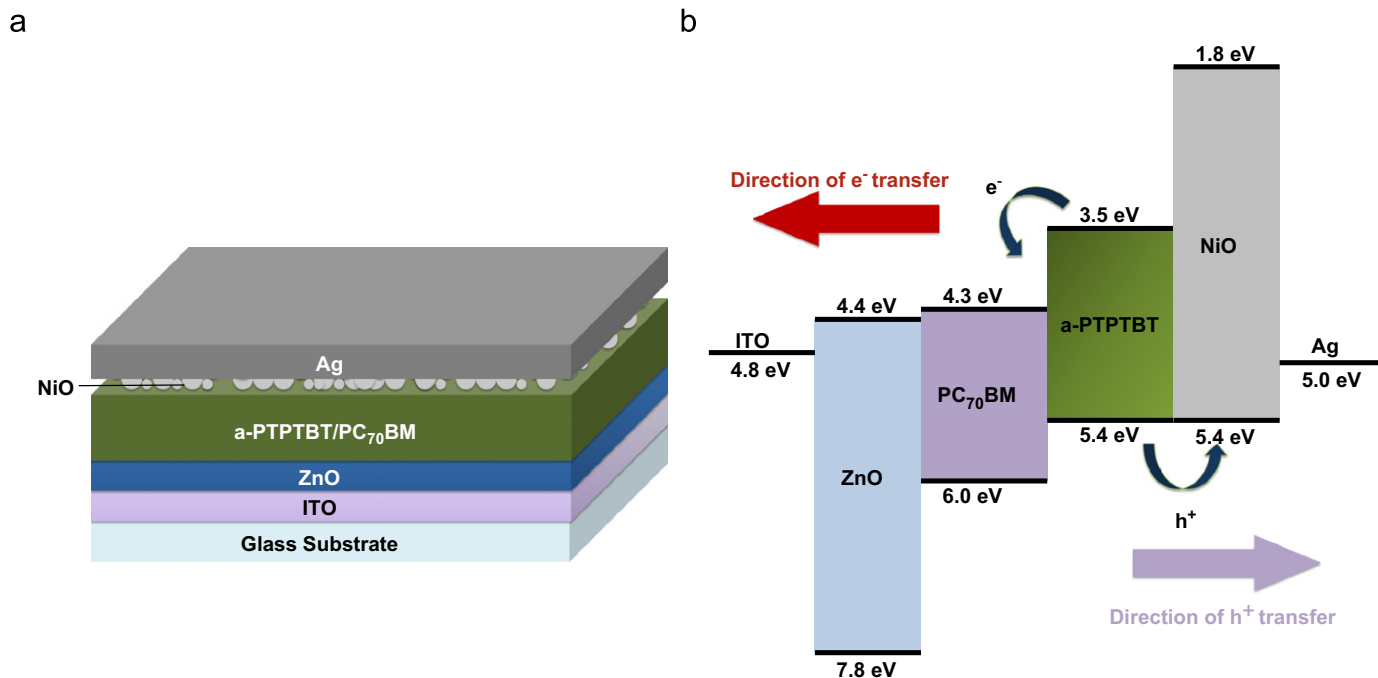


Fig. 2. (a) Device configuration of the inverted a-PTPTBT:PC₇₀BM solar cell. (b) The energy level diagram of the device.

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