

High-efficiency inverted polymer solar cells with solution-processed metal oxides

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ABSTRACT

The selection of carrier transporting layer in polymer solar cells is an important issue because the nature and direction of carrier transport can be manipulated by inserting different functional layers in the device structure. In this work, we report a very efficient inverted polymer solar cell (PSC) system based on regioregular poly(3-hexylthiophene) and a n-type acceptor, bis-indene[C₆₀]. With a pair of metal oxides and the insertion of TiO₂ nanorods electron collecting layer between the ZnO thin film and the active layer, the device efficiency can be greatly improved. The contact area between the active layer and the electron collecting layer, as well as the thickness of active layer, can be increased with the incorporation of TiO₂ nanorods. As a result, photocurrent can be enhanced due to more absorption of light and more charge separation interface. In addition, the larger contact area and the crystalline TiO₂ nanorods provide a more efficient transporting route for the carriers to the cathode. The most efficient device demonstrated shows a high power conversion efficiency of 5.6% with the inverted structure.

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1. Introduction

In recent years, polymer solar cells (PSCs) have emerged as the potential candidate for renewable energy due to the possibility of achieving large area, flexible, and light weight devices with low fabrication cost [1,2]. The bulk-heterojunction PSCs based on regioregular poly(3-hexylthiophene) (rr-P3HT) and fullerene-derivative (6,6)-phenyl C61 butyric acid methyl ester (PCBM) have been reported to reach a power conversion efficiency around 5% [3–5]. The most typical device configuration for this blend system employs an aluminum cathode and a poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS)-modified indium–tin oxide (ITO) or fluorine doped tin oxide (FTO) anode. However, PSCs of this conventional configuration suffer from instability problems. The low work function aluminum is easily oxidized in air, and the acidic PEDOT:PSS would degrade the device performance due to the corrosion to indium–tin oxide (ITO), both of which make the device lifetime very short [6–8]. An approach to solve these problems is to use the inverted structure of polymer

solar cells by modifying the ITO electrode with an electron selecting layer and replacing the easily oxidized low work function metal electrode by a higher work function one to change the nature of carrier transport [9–12]. Device stability utilizing this inverted configuration can be significantly improved [13–15].

Metal oxides can serve as the transporting functional layer in either cathode or anode. Cathode buffer layers such as zinc oxide (ZnO) [11,16], titanium oxide (TiO_x) [12,17], and cesium carbonate (Cs₂CO₃) [10,18,19] have been reported to possess electron selecting and hole blocking functions, and all of them can be solution-processed. On the other hand, metal oxides such as nickel oxide (NiO) [20], vanadium oxide (V₂O₅) [10], and molybdenum oxide (MoO₃) [16] can function as hole transporting and electron blocking layer in the anode side. However, most of these oxides are deposited by vacuum technique, which is rather expensive and therefore not preferable. To further reduce the fabrication cost, solution-processable anode-side metal oxides layer applicable in polymer solar cells have been developed, such as the sol–gel derived copper oxide (CuO_x) film [21] and the interlayer casted by colloidal dispersion of V₂O₅ and WO₃ [15,22]. These solution-processed metal oxides had been discovered to be able to effectively reduce the device leakage current, thereby improving device characteristics. In addition, because of the easy processing with the solution, many conditions could be quickly experimented for optimization. Finally, by sandwiching the organic layer between these metal oxides, direct

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exposure to air can be avoided, resulting in much better device stability.

In this work, we report a very high efficient inverted structure solar cell. It consists of two electron selecting oxide layers, as well as two hole transporting and functional layers, which were all deposited using the solution-process. The ITO electrode is modified by the ZnO sol-gel thin film, and on the top covers the TiO₂ nanorods array. This TiO₂ nanorods layer not only provides larger interfacial area with the organic photoactive blend, but also forms an efficient transporting pathway for electron, thus greatly enhancing the device performance. For hole transport, in addition to the use of solution-processable colloidal suspension of NiO nanoparticles, an additional hole transport ink, developed by Plextronics Inc., is used as the hole transporting layer (HTL) at the anode side. This HTL is based on sulfonated poly(thiophene-3-[2-(2-methoxyethoxy) ethoxy]-2,5-diyl) (S-P3MEET), a stable, inherently doped conjugated polymer. Finally, we demonstrate that inverted PSCs using electronic grade regioregular poly(3-hexylthiophene) and a novel n-type acceptor, bis-indene[C₆₀] (Plexcore[®] PV2000, Plextronics) [23–26] have high power conversion efficiency of 5.6% with $J_{sc}=11.3 \text{ mA/cm}^2$, $V_{oc}=0.79 \text{ V}$, and FF=63% under AM 1.5 G (100 mW/cm²) irradiation intensity when paired with our novel solution-processed metal-oxide interlayer.

2. Experimental

The Plexcore[®] PV2000 photoactive ink and the Plexcore[®] hole transport ink (Plexcore[®] HTL) were provided by Plextronics, Inc. This system has achieved a 5.4%, and later 6.0% power conversion efficiency in conventional device structure and had been certified by National Renewable Energy Laboratory (NREL) [27,28]. The TiO₂ nanorods with diameter of 5 nm and length of 30–40 nm were synthesized by the hydrolysis of titanium tetrakisopropoxide as previously reported [29,30]. Nickel(II) oxide nanopowder (< 50 nm, 99.8%) was purchased from Aldrich and zinc acetate dehydrate was from Sigma-Aldrich. The monoethanolamine (99.5%), the chelating agent and the 2-methoxyethanol (99%), the solvent for sol-gel solution were purchased from Merck and Alfa Aesar, respectively.

The device configuration is shown in Fig. 1. Devices were fabricated on the pre-cleaned ITO coated glass substrate (7 Ω/sq). The ZnO sol-gel thin film (~50 nm) was formed by spin-coating a 0.5 M zinc acetate solution in 2-methoxyethanol and then being annealed at 200 °C for an hour in air. Then TiO₂ nanorods dispersed in pyridine of different concentrations (1.5, 3.0, 3.6, 4.5, and 6.0 mg/ml) were tested and spin-coated onto the ZnO layer at 2000 rpm for 40 s. The samples were heated at 140 °C for 10 min to remove the residual solvent and then transferred into the nitrogen filled glove box. Subsequently, the Plexcore[®] PV2000

photoactive p/n ink was spin-coated on top of TiO₂ nanorods/ZnO layer at 600 rpm for 40 s and slowly dried in a Petri dish for 30 min. Next, the colloidal suspension of NiO nanoparticles (0.1 mg/ml), which served as both electron blocking and hole conducting layer and then the high-hole-mobility Plexcore[®] HTL ink, were spin-coated onto the active layer at 2000 and 4000 rpm, respectively. Finally, silver electrode was deposited by thermal evaporation in a vacuum of 2×10^{-6} torr and the device was upon thermal annealing at 170 °C for 10 min in nitrogen atmosphere.

The devices were unencapsulated and stored in air. The current density–voltage characteristics were measured with a Keithley 2400 source measurement unit. The solar illumination was generated by a Thermo Oriel 150 W solar simulator with AM 1.5 G filters and calibrated at the intensity of 100 mW/cm² by a NREL certified mono-crystalline silicon reference cell (PVM-236). Surface morphology of ZnO layer, together with the TiO₂-nanorods-covered one, was examined by tapping mode atomic force microscopy (NT-MDT Solver P47). A Perkin-Elmer Lambda 35 UV–vis spectrophotometer was used to obtain the device absorption spectra. Field-emission scanning electron microscopy (FESEM) and room temperature photoluminescence spectroscopy (PL) were also applied to analyze device characteristics.

3. Results and discussion

The effects of applying TiO₂ nanorods of different concentration as additional electron collecting network on device performance are investigated. Firstly, all the devices incorporated with TiO₂ nanorods show enhancement of device performance, as shown in Fig. 2. The most apparent improvement of devices lies in V_{oc} (open circuit voltage) and J_{sc} (short circuit current), resulting in substantial improvement in power conversion efficiency (PCE). Secondly, there is an optimal concentration of TiO₂ nanorods. The device fabricated with the 3 mg/ml solution shows the best performance among all the devices, with PCE of 5.6%, V_{oc} of 0.79 V, J_{sc} of 11.3 mA/cm², and fill factor (FF) of 63%. As the concentration of TiO₂ solution increased from 1.5 to 3.0 mg/ml, the ZnO surface can be covered more evenly and fully by the nanorods. Therefore, the device performance can be improved. However, when the concentration of TiO₂ nanorods further increases, nanorods start to pile up unevenly onto the ZnO layer, resulting in increase of the contact resistance and thus leading to a lower photocurrent and PCE. To sum up, the above results suggest that adding small amount of TiO₂ nanorods between the ZnO and active layer in inverted structure can serve as an effective method to improve the device efficiency, and the details of the device performance are summarized in Table 1. The average value (solid squares) and standard deviation (error bars) of the parameters were derived

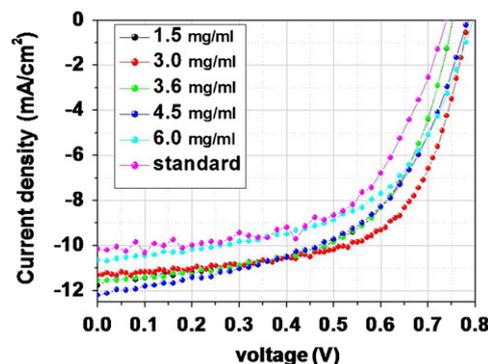


Fig. 2. J - V characteristics of devices with different concentrations of TiO₂ nanorods under 100 mW/cm² AM 1.5 G light illumination. All the devices incorporated with TiO₂ nanorods show enhancement of device performance and the optimal concentration of TiO₂ nanorods is 3 mg/ml.

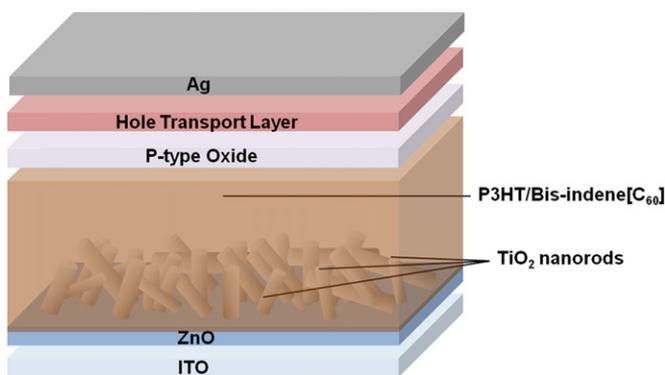


Fig. 1. Schematic diagram of inverted-type device configuration incorporating TiO₂ nanorods as additional electron collecting layer.

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