



Enhanced photovoltaic performance of inverted polymer solar cells by tuning the structures of titanium dioxide



Ruixiang Peng^a, Feng Yang^a, Xinhua Ouyang^a, Ying Liu^a, Yong-Sang Kim^b, Ziyi Ge^{a,*}

^a Ningbo Institute of Material Technology & Engineering, Chinese Academy of Sciences, Ningbo 315201, PR China

^b Department of Electrical Engineering, Myongji University, Gyeonggi 449-728, South Korea

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ABSTRACT

Inverted polymer solar cells using TiO₂ film as electron transporting layer were fabricated with the structure of fluorine-doped tin oxide/TiO₂ films/poly(3-hexylthiophene): [6,6]-phenyl-C₆₁-butyric acid methyl ester/Poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate)/Ag. By tuning the crystalline structure of TiO₂ films, the photovoltaic performance of the devices was remarkably enhanced. TiO₂ was prepared by sol-gel method and their structures, morphologies and transmittance were characterized by X-ray diffraction, scanning electron microscope, and UV-visible spectrophotometer. Interestingly, for TiO₂-3 film, which was prepared with tetrabutyl titanate, acetyl acetone and ethanol in a ratio of 1:0.5:6, the open-circuit voltage and fill factor of the device were up to 0.6 V and 64.8%, respectively, and the power conversion efficiency of TiO₂-3 film was achieved up to 3.56% with the current density of 9.18 mA/cm² under an AM 1.5 G (100 mW/cm²) irradiation intensity. In the meanwhile, the stabilities of these devices were also studied and results showed that our work was better than the corresponding devices of conventional structure.

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1. Introduction

Bulk heterojunction solar cells have been attracting extensive research efforts in the past decades due to their advantages of light-weight, low cost and flexibility. The performance of laboratory-scale polymer solar cells based on different electron donor and acceptor materials have been remarkably improved, and some reports with power conversion efficiencies up to 7% at Air Mass 1.5 Global (AM 1.5G) has been demonstrated [1–3]. In these reports, the typical cells consist of a bottom indium tin oxide (ITO) anode, a Poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS) anode interfacial layer, a photo-active layer and a low-work-function top metal cathode. However, studies showed that PEDOT:PSS degraded the property of devices due to its corrosion of ITO [4]. In addition, the prolonged exposure of these kinds of devices to air can lead to oxidation of the electrode and degradation of the active layer from oxygen and moisture diffusion through grain boundaries and pinholes of the metal electrode without encapsulation, which will seriously limit their application [5,6]. In order to solve these problems, an approach was employed by inserting a metal oxide buffer layer between the active layer and ITO anode to reduce the amount of damage and oxygen diffusion into the polymer. Recently, different types of solution-processed sol-gel titanium oxide and zinc oxide

have been used as buffer layer to overcome these problems and improve device stability in ambient conditions, these architectures are called “inverted cells” [7–11]. Nanocrystalline TiO₂ porous films are widely used for the dye-sensitized solar cells, this type of solar cells have achieved the overall efficiency of 10% and remarkable stability under simulated solar light [12,13], which is comparable to that of amorphous silicon solar cells. TiO₂ films also provide a promising alternative to the electron transporting layer because of their low work function, high electron mobility and optical transparency, as well as their ease of synthesis [14,15]. Despite these advantages, power conversion efficiencies of most of the TiO₂ based inverted polymer solar cells with poly(3-hexylthiophene): [6,6]-phenyl-C₆₁-butyric acid methyl ester (P3HT:PCBM) as active layer were below 3% [7,16]. TiO₂ film plays a significant role to affect the performance of this type of solar cells, the major challenges in using TiO₂ as ETL include poor spatial distribution of the nanoparticles over a large area [17,18]. Accordingly, it is necessary to optimize TiO₂ films structure so as to realize high efficiency inverted polymer solar cells.

In order to deeply improve the power conversion efficiency (PCE) of inverted polymer solar cells (PSCs), we synthesized a series of crystalline TiO₂ films in this paper. Their structures and morphologies were characterized by X-ray diffraction (XRD) and Scanning electron microscopy (SEM). The transmittance of TiO₂ substrates were tested by UV-visible spectrophotometer (UV-vis). The inverted polymer solar cells using TiO₂ films as electron transporting layers were fabricated with the following architecture: Fluorine-doped tin oxide (FTO)/TiO₂/P3HT:PCBM/PEDOT:PSS/Ag. In addition, the relationship between

* Corresponding author. Tel./fax: +86 574 86680273.
E-mail address: geziyi@nimte.ac.cn (Z. Ge).

the performance of the photovoltaic cells and TiO₂ thin film structures were demonstrated in the article.

2. Experimental details

2.1. Materials and the methods

Fluorine-doped tin oxide glass sheet (2.2 mm thick, $\leq 14 \Omega/\text{square}$, transmittance > 90%) was purchased from Nippon Sheet Glass Company, Ltd, PEDOT:PSS (Baytron® P VP Al 4083) and PC₆₀BM were purchased from Luminescence Chemical Engineering Technology Co., Ltd, titanium tetraisopropoxide (Acros, 98 +%), titanium isopropoxide (Acros, 98 +%), tetrabutyl titanate (Acros, 98 +%), titanium ethoxide (Acros, 98 +%), dichlorobenzene and hexamethylene disilazane (HMDS) were purchased from Acros. All other reagents were obtained from Sinopharm Chemical Reagent Co., Ltd, and used as received if not specified. P3HT was synthesized following the method described elsewhere [19], the average molecular mass (Mw) of the synthesized polymer was 32,000 with a polydispersity index of 1.6, as determined with a WATERS 150-C gel permeation chromatography (GPC) with THF as solvent and mono-dispersed polystyrene as standard.

2.2. Preparation of TiO₂ solutions

Four different kinds of TiO₂-sol were prepared and named TiO₂-1, TiO₂-2, TiO₂-3 and TiO₂-4.

TiO₂-1 Titanium tetraisopropoxide, absolute ethanol, and acetic acid were mixed with the volume ratio of 1:20:0.1. The reaction was stirred at room temperature until a homogeneous colorless stable TiO₂-sol was obtained, which was assigned as TiO₂-1.

TiO₂-2 Titanium isopropoxide (C₁₂H₂₈O₄Ti), isopropanol, hydrochloric acid (HCl, 0.28 mol/L), and distilled water were mixed with the volume ratio of 3:20:0.1:0.2. The reaction was stirred at room temperature until a yellowish color TiO₂-sol was formed and named as TiO₂-2.

TiO₂-3 The preparation process was same with the TiO₂-1. We changed the Titanium tetraisopropoxide to tetrabutyl titanate (Ti(OC₄H₉)₄) and the ratio of 1:0.5:6 to Ti(OC₄H₉)₄, acetyl acetone and ethanol in the reaction. A stable yellow TiO₂-sol was synthesized and labeled as TiO₂-3.

TiO₂-4 The preparation process was similar as above. TiO₂ was prepared by mixing titanium (IV) ethoxide, HCl (0.28 mol/L), and isopropyl alcohol with the ratio of 5:1:30, a light yellow stable TiO₂-sol was obtained and labeled as TiO₂-4.

2.3. Devices fabrication

A solution of P3HT and PCBM was prepared using 1,2-dichlorobenzene as solvent. The concentration of the solution was maintained around 35 mg/mL with 1% (v/v) 1,8-octanedithiol. The inverted polymer solar cells with the sandwich structure of FTO/TiO₂/P3HT:PCBM/PEDOT:PSS/Ag were fabricated. The above-prepared TiO₂-sol was spin-coated on a cleaned FTO substrates (2.54 cm × 2.54 cm), which were cleaned by a routine cleaning procedure which includes initial manual washed in aqueous detergent, and then sequentially sonication in acetone, isopropanol, and deionized water, lastly rinsing in ethanol and drying in a N₂ stream. The TiO₂ layer was sintered at 500 °C for 1 h under ambient conditions. The pre-dissolved composite solution was filtered through 0.45 μm syringe filter and then an active layer of about 300 nm was spin coated on the TiO₂ layer. Subsequently, hexamethyldisilazane (HMDS) was coated immediately onto the active layer. PEDOT:PSS mixed with 1% (v/v) of Triton X-100 (C₁₄H₂₂O(C₂H₄O)_n) nonionic surfactant was spin-coated onto the surface of the active layer. Thermal preannealing was conducted at 120 °C for 30 min on a hot plate in a glovebox (H₂O ≤ 0.1 ppm, O₂ ≤ 0.1 ppm). Finally, a

cathode (top electrode) of Ag was deposited onto the PEDOT:PSS layer in a thermal evaporator under a vacuum of 5×10^{-5} Pa. The effective area of single solar cells was 0.1257 cm². The device structure of FTO/TiO₂ films/P3HT:PCBM/PEDOT:PSS/Ag is shown in Fig. 1.

2.4. Characterization

The thickness of films was measured using surface profiler (Dektak 150). The transmittance spectra of FTO and TiO₂ films were recorded using a Lambda 950 spectrophotometer. The crystalline structures were characterized by XRD (D8 advance) with a 2θ from 15° to 80° by 0.02° s⁻¹ steps operating at 40 kV accelerating voltage and 40 mA current using Cu K radiation source, the incident angle was kept constant at 0.5° throughout the experimentation. The surface morphology and pore distribution of the produced films were studied using SEM at 4.0 kV (FEI Quanta FEG 250). The current–voltage (I–V) characteristics of all the photovoltaic cells were measured under the simulated solar light (100 mW cm⁻²; AM 1.5 G) provided by a Newport-Oriel® Sol3A 1000-W solar simulator and stored in Ar atmosphere without encapsulation. Electrical data were recorded using a Keithley 2440 source-measure unit, the intensity of the simulated solar light was calibrated by a standard Si photodiode detector (PV measurements Inc.), which was calibrated at National Renewable Energy Laboratory of the United-States. The external quantum efficiency (EQE) spectra were measured by a Newport-Oriel® IQE 200™ under illumination with monochromatic light from a Xe lamp at room temperature in Air atmosphere.

3. Results and discussion

Fig. 2 shows the surface micrographs of different TiO₂ films using SEM which were spin-coated on the FTO substrate and annealed at 500 °C. As shown in the figures, TiO₂-1 film (a) presented a smooth morphology in elongated domains, but in the edge area, the surface with many cracks was observed. For the substrate of TiO₂-2, the surface is inhomogeneously covered by large aggregates consisting of typically 5 μm dimension scales, the aggregates are separated by many cracks probably formed during the drying process due to surface tension between the film and the air [20]. The large aggregates in the matrix may limit the transformation from anatase to rutile phase, and the sintering process resulted in a low crystallite and a poor charge mobility of the film. In picture (c) TiO₂-3 films and (d) TiO₂-4 films, well-defined homogeneous surface morphologies with nanoparticle-shaped domains can be clearly seen. These equally distributed nanoparticles can offer more interface for charge separation and transmission between the inorganic TiO₂ composites and donor materials, which will be beneficial to enhance the performance of solar cells.

To reveal the deep information of the crystal nanoparticle's size and structure, TiO₂ films were studied by X-Ray Diffraction measurement, which can be used more quantitatively to study particle size and distribution of nanoparticles as well as other structure-related parameters

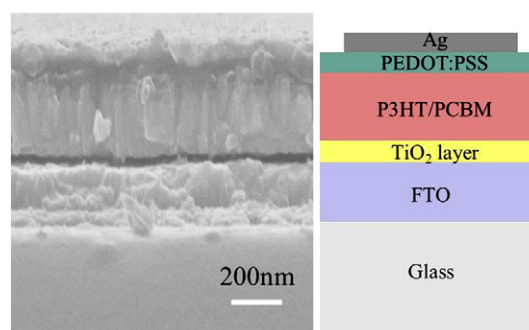


Fig. 1. SEM image of device cross section and schematic structure of the inverted polymer solar cells.

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