



Simple solution-processed titanium oxide electron transport layer for efficient inverted polymer solar cells

Liang Sun^{a,c}, Wenfei Shen^{a,b}, Weichao Chen^a, Xichang Bao^{a,*}, Ning Wang^a, Xiaowei Dou^a, Liangliang Han^a, Shuguang Wen^a

^a CAS Key Laboratory of Bio-based Materials, Qingdao Institute of Bioenergy and Bioprocess Technology, Chinese Academy of Sciences, Qingdao 266101, China

^b Institute of Hybrid Materials, Laboratory of New Fiber Materials and Modern Textile—The Growing Base for State Key Laboratory, Qingdao University, Qingdao 266071, China

^c University of Chinese Academy of Sciences, Beijing 100049, China

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ABSTRACT

Titanium oxide (TiO_x) is an effective electron transport layer (ETL) in polymer solar cells (PSCs). We report efficient inverted PSCs with a simple solution-processed amorphous TiO_x (s- TiO_x) film as an ETL. The s- TiO_x film with high light transmittance was prepared by spin-coating titanium (IV) isopropoxide isopropanol solution on indium tin oxide coated glass in inert and then placed in air under room temperature for 60 min. The introduction of s- TiO_x ETL greatly improved the short circuit current density of the devices. PSCs based on poly(3-hexylthiophene):[6,6]-phenyl-C61-butyric acid methyl ester and poly(4,8-bis-alkyloxy-benzo[1,2-b:4,5-b']dithiophene-alt-alkylcarbonyl-thieno[3,4-b]thiophene):[6,6]-phenyl-C71-butyric acid methyl ester using s- TiO_x film as ETL shows high power conversion efficiency of 4.29% and 6.7% under the illumination of AM 1.5G, 100 mW/cm², which shows enhancements compared to the conventional PSCs with poly(styrenesulfonate)-doped poly(ethylenedioxythiophene) as anode buffer layer. In addition, the device exhibits good stability in a humid ambient atmosphere without capsulation. The results indicate that the annealing-free, simple solution processed s- TiO_x film is an efficient ETL for high-performance PSCs.

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1. Introduction

Bulk heterojunction polymer solar cells (PSCs) have been widely studied owing to the growing need for renewable energy. The most highlighted attribute of PSCs is the potential for light weight, flexibility, solution processing, and low cost large-scale production [1–5]. With great research efforts over the past decade, the power conversion efficiencies (PCEs) for single-junction cells over 9% [6] and tandem cells over 10% [7,8] have been reported. The device performance improvements not only strongly based on the use of donor and acceptor materials, but also related to the functional interface materials, which can greatly affect charge transport, collection, and consequently the PCEs of the devices [6,9–11].

In conventional PSCs, poly(styrenesulfonate)-doped poly(ethylenedioxythiophene) (PEDOT:PSS) is the widely used hole transport layer. However, its acidity, tendency to moisture absorption, and inability to block electrons effectively, together with the low-work-function metal cathode are the factors which contribute to device performance problems and degradation [12–14]. The inverted PSCs with air-stability, high-work-function metals (Au, Ag) covering on p-type metal oxide interface layers (molybdenum oxide, MoO_3 [15], tungsten oxide, WO_3 [16], vanadium

oxide, V_2O_5 [17]) as top anode, and stable n-type metal oxides such as titanium oxide (TiO_x) [18,19], zinc oxide (ZnO) [20,21] modified indium tin oxide (ITO) coated glass as the cathode, could solve the stability problem of conventional solar cells.

Titanium oxide is a suitable interface material because of its wide-gap and optical transparency, as well as high electron mobility. However, the typical crystalline TiO_2 usually requires a high-temperature treatment (above 450 °C) by sol–gel techniques, which restricts its application in the large-scale production [22,23]. To avoid high annealing temperature, the amorphous TiO_x films, fabricated by a solution-based sol–gel process have been reported [20,24,25]. The amorphous TiO_x films have many advantages, such as low-temperature process, excellent stability, inexpensive, non-toxicity, and compatibility with flexible plastic processing at room temperature. However, there are some shortcomings, such as preparing the amorphous TiO_x nanoparticles first and then preparing the solution to be used. The used TiO_x precursor solution obtained by the reaction of titanium (IV) isopropoxide (TTIP) and other compositions firstly. The core reaction for preparation of TiO_x is the hydrolysis of TTIP. Since TTIP can quite easily react with water and convert to titanium oxide, it is feasible to form amorphous TiO_x thin film in a very simple way via direct hydrolysis of TTIP thin film in air instead of precursor solution.

In this article, one step solution-processed amorphous TiO_x (named s- TiO_x) films were prepared by spin coating TTIP isopropanol solution

* Corresponding author. Tel.: +86 532 80662701; fax: +86 532 80662778.
E-mail address: baoxc@qibebt.ac.cn (X. Bao).

on ITO coated glass in glovebox filled with N_2 , and then the TTIP films were placed in air for hydrolysis. The TTIP can easily decompose into amorphous TiO_x and the $s-TiO_x$ films with high transmittance are very smooth. We studied the photovoltaic performance of the $s-TiO_x$ electron transport layer by fabricating the PSCs. The molecular structures of the donor materials (poly(3-hexylthiophene) (P3HT) and poly(4,8-bis-alkyloxy-benz[1,2-b:4,5-b']dithiophene-alt-alkylcarbonyl-thieno[3,4-b]thiophene) (PBDDTTT-C)) and acceptor materials ([6,6]-Phenyl-C61-butyric acid methyl ester ($PC_{61}BM$) and [6,6]-phenyl-C71-butyric acid methyl ester ($PC_{71}BM$)) are illustrated in Fig. 1a. The PCEs of the PSCs based on P3HT: $PC_{61}BM$ and PBDDTTT-C: $PC_{71}BM$ with $s-TiO_x$ electron transport layer anode buffer layer reach 4.29% and 6.7% under the illumination of AM 1.5G 100 mW/cm^2 , respectively. The improved PCE mainly attributed to the suitable work function of $s-TiO_x$ film and the enhanced electron collection of the active layer in the devices. The results demonstrate that single step proceed $s-TiO_x$ is a promising choice for high-efficiency inverted PSCs and for flexible devices.

2. Experimental sections

2.1. Materials and fabrication of PSCs

Patterned ITO coated glass with a sheet resistance of 15 Ω/sq was purchased from Shenzhen Display (China). P3HT and PBDDTTT-C were purchased from Lumtec and Solarmer Inc., respectively. $PC_{61}BM$ and $PC_{71}BM$ were obtained from American Dye Sources (ADS). Titanium (IV) isopropoxide (TTIP) was purchased from Aladdin (AR grade, China). All reagents were used as received without further purification. Eight types of devices with different structures were designed to

investigate the effect of the $s-TiO_x$ interface layer on the performance of the PSCs. The structures: (A) ITO/ $s-TiO_x$ /P3HT: $PC_{61}BM$ /MoO₃/Ag; (B) ITO/PEDOT:PSS/P3HT: $PC_{61}BM$ /Ca/Al; (C) ITO/ $s-TiO_x$ (in air)/P3HT: $PC_{61}BM$ /MoO₃/Ag; (D) ITO/ TiO_2 nanoparticle film/P3HT: $PC_{61}BM$ /MoO₃/Ag; (E) ITO/ZnO/P3HT: $PC_{61}BM$ /MoO₃/Ag; (F) ITO/P3HT: $PC_{61}BM$ /MoO₃/Ag; (G) ITO/ $s-TiO_x$ /PBDDTTT-C: $PC_{71}BM$ /MoO₃/Ag; and (H) ITO/PEDOT:PSS/PBDDTTT-C: $PC_{71}BM$ /Ca/Al. Here, TiO_2 nanoparticles (NPs, with sizes less than 7 nm) film was about 30 nm thick with a root mean square (RMS) less than 2.5 nm was prepared as described by Yang et al. [26]. ZnO film (with sizes of about 9 nm) was about 40 nm thick with a RMS of about 2 nm obtained as we stated before [27]. The $s-TiO_x$ here was prepared by spin coating (3000 rpm) 5% v/v TTIP isopropanol solution on the ITO coated glass in glovebox filled with N_2 , and then was exposed in air for 60 min at room temperature. In comparison, another sample $s-TiO_x$ (in air) was prepared by spin coating in air and treated as above. Subsequently, the $s-TiO_x$ films were transferred to glovebox. After cleaning ITO coated glass with detergent, deionized water, acetone and isopropanol in an ultrasonic bath for 20 min each time, the ITO coated glass was exposed to oxygen plasma to remove organic contaminants and increase the wetting envelope. Then, the above mentioned electrode interface layers were prepared and treated. Following that, the active layer was spin-coated on them. The P3HT: $PC_{61}BM$ (1:1 w/w, polymer concentration of 18 mg/mL in chlorobenzene) solution were spin coated at 600 rpm, and baked at 150 $^{\circ}C$ for 10 min, respectively. The active layer of PBDDTTT-C: $PC_{71}BM$ is the same as that described in Ref. [28]. Finally, MoO₃ (5 nm)/Ag (80 nm) or Ca (10 nm)/Al (100 nm) layer was evaporated for inverted or conventional devices. The device area was 0.1 cm^2 defined by the shadow mask.

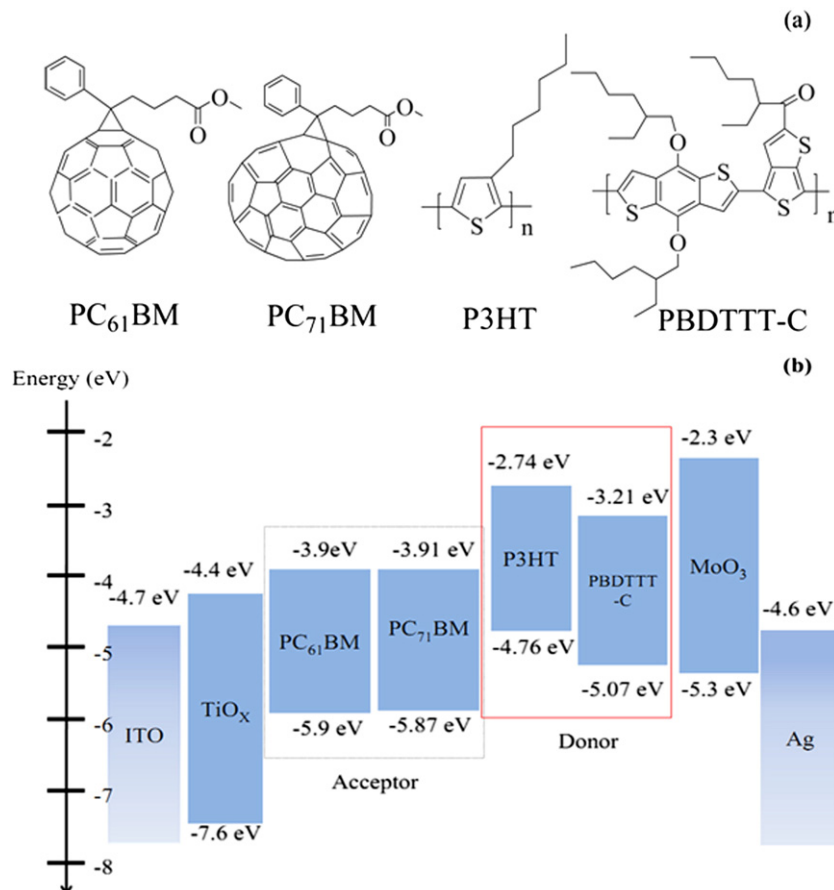


Fig. 1. (a) The molecular structures of $PC_{61}BM$, $PC_{71}BM$, P3HT, and PBDDTTT-C. (b) The schematic energy diagram of the materials involved in the PSCs.

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