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Light illumination intensity dependence of current-voltage characteristics in polymer solar cells with solution-processed titanium chelate as electron extraction layer

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ABSTRACT

Solution-processed titanium (IV) oxide bis(2,4-pentanedionate) (TOPD) as an electron extraction layer (EEL) and the blend of thieno[3,4-b]thiophene/benzodithiophene (PTB7) and [6,6]-phenyl C71-butyric acid methyl ester (PC₇₁BM) as a photoactive layer with a power conversion efficiency (PCE) of 9.15% are demonstrated. Analysis of the current-voltage (J-V) characteristics under dark conditions provides information on the weaker leakage current of TOPD-based polymer solar cells (PSCs) compared to ZnO-based PSCs. By investigating the photovoltaic performance under different light illumination intensities, we achieved an improvement of photovoltaic performance under weak light illumination intensity for TOPD-based PSCs. The enhanced photovoltaic performance under weak light illumination intensity for TOPD-based PSCs can be experimentally explained by the higher open-circuit voltage (V_{OC}) value under lower light illumination intensity compared to ZnO-based PSCs. This indicates that the TOPD layer in the PSCs reduced the probability of charge carrier recombination at trap sites and improved the charge carrier collection and transport efficiency under weaker light illumination intensity. Simultaneously, the similar fill factor and short circuit current density of two PSCs can be experimentally explained by the weak bimolecular recombination between the photoactive layer and indium-tin-oxide electrode with the different EEL materials under different light illumination intensity.

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1. Introduction

Solution-processable polymer solar cells (PSCs) have attracted much attention owing to their lightweight, flexible, low-cost, and roll-to-roll fabrication properties ([Dominici et al., 2013; Tang,](#page--1-0) [1986\)](#page--1-0). Significant efforts are now focused on how to further enhance their power conversion efficiency (PCE), which has recently exceeded 12% for ternary PSCs [\(Zhao et al., 2016; Li](#page--1-0) [et al., 2016](#page--1-0)). Regular PSCs have a sandwich structure with corrosive and hygroscopic poly(3,4-ethylenedioxylenethiophene):poly(styre nesulfonic acid) (PEDOT:PSS) as a hole extraction layer (HEL), Ca as an electron extraction layer (EEL), and Al as a cathode [\(Liu](#page--1-0) [et al., 2014; Oh et al., 2015; Marinova et al., 2017\)](#page--1-0). PEDOT:PSS has a corrosive and hygroscopic nature and Ca is active and can be easily oxidized by oxygen and water, which is detrimental to device lifetime and limits its application. The development of new EEL materials with a noncorrosive, solution-processable, and low-temperature thermal annealing treatment is critical to obtaining high-performance PSCs [\(Wang et al., 2015; Yin et al., 2016\)](#page--1-0).

ZnO nanoparticles have been synthesized following the Pacholski method [\(Gonzalez-Valls and Lira-Cantu, 2009](#page--1-0)) and used as an EEL material for common inverted PSCs. They have the advantages of low cost and easy solution-processability. However, ZnO nanoparticle aggregation will occur over 12 h using this recipe in our experimental process. Titanium oxide (TiO_X) is one of the most promising materials for this application ([Gregg, 1996; Kim et al.,](#page--1-0) [2006\)](#page--1-0), which transports electrons from the energy level of the lowest unoccupied molecular orbitals (E_{LUMO}) of the acceptor materials to the cathode. However, the common fabrication method of a TiO_x layer is vacuum deposition, which has the disadvantages of high cost and limited large-scale production, among others. Thus, the design and synthesis of solution-processed TiO_X as an anode buffer layer in PSC devices has attracted the interest of many research groups. Yongfang et al. reported that inverted PSCs were fabricated

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with the titanium chelate, titanium di-isopropoxide bis(2,4 pentanedionate) (TIPD) as an EEL by spin-coating a TIPD isopropyl alcohol solution on the top of indium-tin-oxide (ITO) glass [Tan](#page--1-0) [et al., 2012; Wang et al., 2012, 2013,](#page--1-0) which has the advantages of solution processability, stable chemical properties, etc. In this study, we introduced the aforementioned alcohol-soluble titanium chelate, TIPD, as an EEL to replace ZnO nanoparticles ([Wang et al.,](#page--1-0) [2013; Tan et al., 2012\)](#page--1-0). After the thermal treatment of the TIPD layer, two isopropoxy groups of TIPD molecules were hydrolyzed and formed $Ti=O$ bonds and a titanium (IV) oxide bis(2,4pentanedionate) (TOPD) layer [Tan et al., 2012; Yang et al., 2014.](#page--1-0) The chemical structures of TIPD and TOPD are shown in Fig. 1 (a) and (b), respectively. Thus, TIPD is suitable for the EEL between the photoactive layer and electrode. The TIPD solution and TOPD layer possess the advantages of low cost, stable chemical characteristics, and low thermal annealing temperature (130 \degree C). Furthermore, for the TOPD layer fabrication, the solution process using isopropanol solvent and water could be carried out with common technologies, such as the spin-coating method ([Tan et al., 2012;](#page--1-0) [J](#page--1-0)ø[rgensen et al., 2008](#page--1-0)). The blend of thieno[3,4-b]thiophene/benzo dithiophene (PTB7) and [6,6]-phenyl C71-butyric acid methyl ester $(PC_{71}BM)$ acts as a photoactive layer, and the composite layer of MoO3/Ag acts as a anode. The inverted PSC structure is ITO/ TOPD/PTB7: PC_{71} BM/MoO₃/Ag. Over the course of an entire day, the light illumination intensity of most light illuminated time is lower than 1 Sun or 100 mW cm^{-2} . Thus, photovoltaic perfor-

mance as a function of light illumination intensity is important ([Corazza et al., 2015; del Pozo et al., 2017](#page--1-0)). Photovoltaic performance as a function of light illumination intensity was measured under illumination using an AM 1.5G solar simulator, and the light illumination intensity changed between 10 and 100 mW cm⁻². At the same time, we studied the effect of TOPD as an EEL compared to ZnO as an EEL on photovoltaic performance at weak light illumination intensity. We also studied the recombination dynamics of PSCs by analyzing the change of photovoltaic performance parameter (*Ji.e., open-circuit voltage (* V_{OC} *), short-circuit current density* (I_{SC}) , fill factor (FF), and power conversion efficiency (PCE)] dependence on light illumination intensity.

2. Experimental details

2.1. Materials

PTB7 was purchased from Solarmer Materials, Inc.; PC_{71} BM from Nano.C; TIPD, isopropyl alcohol, o-dichlorobenzene (ODCB, anhydrous, 99%), and 1,8-di-iodooctane (DIO) from Sigma-Aldrich Co.; and $MoO₃$ and Ag from Alfa Aesar Co. ZnO solution was synthesized by the sol-gel method [\(Mikroyannidis et al., 2009\)](#page--1-0).

2.2. Device preparation and characteristics

The TIPD solution was prepared by adding 0.4 g of TIPD into 10 mL of organic solvent (the ratio of isopropanol solvent and deionized water is 4:1), and the solution was stored for 2 h in air. PTB7: PC_{71} BM (1:1.5, wt.%) blends were dissolved in an ODCB solution overnight, with a PTB7 concentration of 10 mg m l^{-1} . To obtain better photovoltaic results, 3% [1,8-di-iodooctane (DIO)/1,2 dichlorobenzene (DCB), v/v] of DIO as an additive in PTB7:PC₇₁BM blend solution. The ITO substrates were ultrasonicated in 30 \degree C aqueous detergent followed by isopropyl alcohol, acetone, and deionized water for 30 min each. Subsequently, the substrates were dried under a stream of nitrogen and hot-stage heating. UV-ozone treatment of the ITO glasses was then carried out for 20 min.

Fig. 1. Chemical structure of (a) TIPD and (b) TOPD and (c) PSC configuration and (d) energy band diagram of the inverted PSCs with TOPD and ZnO as an EEL and PTB7: PC₇₁BM as a photoactive layer.

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