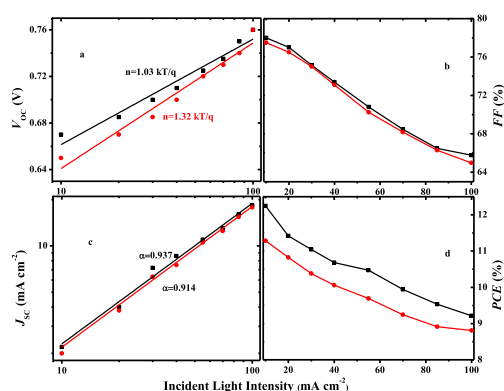


Regular Article

Light illumination intensity dependence of photovoltaic parameter in polymer solar cells with ammonium heptamolybdate as hole extraction layer

Zhiyong Liu^b, Shengli Niu^{a,*}, Ning Wang^{c,*}^a Key Laboratory of Zoonosis of Liaoning Province, School of Animal Science and Veterinary Medicine, Shenyang Agricultural University, Shenyang 110866, People's Republic of China^b College of Science, Shenyang Agricultural University, Shenyang 110866, People's Republic of China^c Key Laboratory of Physics and Technology for Advanced Batteries (Ministry of Education), College of Physics, Jilin University, Changchun 130012, People's Republic of China

GRAPHICAL ABSTRACT



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ABSTRACT

A low-temperature, solution-processed molybdenum oxide (MoO_x) layer and a facile method for polymer solar cells (PSCs) is developed. The PSCs based on a MoO_x layer as the hole extraction layer (HEL) is a significant advance for achieving higher photovoltaic performance, especially under weaker light illumination intensity. Fourier transform infrared spectroscopy (FTIR) and X-ray photoelectron spectroscopy (XPS) measurements show that the $(\text{NH}_4)_6\text{Mo}_7\text{O}_{24}$ molecule decomposes and forms the molybdenum oxide (MoO_x) molecule when undergoing thermal annealing treatment. In this study, PSCs with the MoO_x layer as the HEL exhibited better photovoltaic performance, especially under weak light illumination intensity (from 100 to 10 mW cm^{-2}) compared to poly(3,4-ethylenedioxythiophene):polystyrene sulfonate (PEDOT:PSS)-based PSCs. Analysis of the current density-voltage (J - V) characteristics at various light intensities provides information on the different recombination mechanisms in the PSCs with a MoO_x and PEDOT:PSS layer as the HEL. That the slopes of the open-circuit voltage (V_{oc}) versus light illumination intensity plots are close to 1 unity (kT/q) reveals that bimolecular recombination is the dominant and weaker monomolecular recombination mechanism in open-circuit conditions. That the slopes of the short-circuit current density (J_{sc}) versus light illumination intensity plots are close to 1 reveals that the effective charge carrier transport and collection mechanism of the MoO_x /indium tin oxide (ITO)

* Corresponding authors.

E-mail addresses: niushengli1@163.com (S. Niu), eningwang@outlook.com (N. Wang).

anode is the weaker bimolecular recombination in short-circuit conditions. Our results indicate that MoO_x is an alternative candidate for high-performance PSCs, especially under weak light illumination intensity.

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1. Introduction

Polymer solar cells (PSCs) have attracted increasing attention as a renewable energy source in recent years [1–3]. Significant efforts are now focused on further enhancing their power conversion efficiency (PCE), which has recently exceeded 12% for ternary PSCs [4,5]. Regular PSCs have a sandwich structure with corrosive and hygroscopic poly(3,4-ethylenedioxyethiophene):poly(styrene sulfonic acid) (PEDOT:PSS) as the hole extraction layer (HEL), Ca as the electron extraction layer (EEL), and a low-work-function metal such as Al as the cathode [6,7]. Ca is active and can be easily oxidized by oxygen and water [8,9], which is detrimental to device lifetime and limits its applications. Owing to their low cost and stable nature, metal oxides (V_2O_5 [10], MoO_3 [11], and WO_3 [12]) have been used as the HEL to replace PEDOT:PSS. Because of its non-acidic nature and deeper energy level of highest occupied molecular orbital (E_{HOMO}), MoO_3 is one of the most promising materials for fabricating PSCs. However, the required vacuum deposition method has limitations in large-scale production [11,13]. Thus, the design and synthesis of solution-processed MoO_3 as an anode buffer layer in PSC devices has attracted the interest of many research groups [14–16].

Yang has reported that the $(\text{NH}_4)_6\text{Mo}_7\text{O}_{24}\cdot 4\text{H}_2\text{O}$ molecule decomposes and forms the molybdenum oxide (MoO_x) molecule when subjected to lower-temperature annealing treatment (optimized thermal annealing temperature and times equal to or greater than 100°C and 10 min, respectively) [17]. In the work, we fabricated a MoO_x layer by spin-coating $(\text{NH}_4)_6\text{Mo}_7\text{O}_{24}\cdot 4\text{H}_2\text{O}$ solution on the top of indium tin oxide (ITO) glass, followed by thermal annealing treatment. A blend of thieno[3,4-*b*]thiophene/benzodithiophene (PTB7) and [6,6]-phenyl C71-butyric acid methyl ester (PC_{71}BM) acts as a photoactive layer. In order to avoid the oxidizing reaction of Ca by oxygen and water in the air, the conjugated polyelectrolyte poly[9,9-bis(3'-(*N,N*-dimethylamino)-propyl)-2,7-fluorene]-*alt*-2,7-(9,9-dioctylfluorene)] (PFN) layer is fabricated as an EEL by spin-coating PFN solution on top of the photoactive layers to replace Ca, and the composite layer of PFN/Al acts as a cathode. The PSC structure is ITO/ MoO_x /PTB7: PC_{71}BM /PFN/Al. Over the course of an entire day, the light illumination intensity during the time the PSC was illuminated by the most light was lower than 100 mW cm^{-2} . Thus, photovoltaic performance as a function of light illumination intensity is important [18,19]. At the same time, the open-circuit voltage (V_{OC}) and short-circuit current density (J_{SC}) as a function of light illumination intensity was measured to gain deeper insight into the charge carrier recombination kinetics among the photoactive layer, ITO modified layer and ITO anode. The photovoltaic parameters [V_{OC} , J_{SC} , fill factor (FF), and PCE] as functions of light illumination intensity were measured under illumination using an AM 1.5G solar simulator, and the light illumination intensity changed between 10 and 100 mW cm^{-2} . The control PSC device with a PEDOT:PSS layer as the HEL had the same configuration. We also studied the recombination dynamics of PSCs by analyzing the change of photovoltaic performance parameter dependence on the light illumination intensity. The PSC configuration and energy-band diagram are shown in Fig. 1(a) and (b); the energy levels of the highest occupied molecular orbitals (E_{HOMO}) and those of the lowest unoccupied molecular orbitals (E_{LUMO}) of all material were obtained from the literature [20,21].

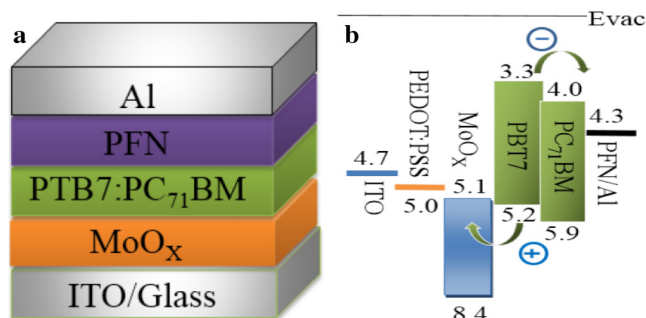


Fig. 1. (a) Configuration and (b) energy-band diagram of the PSCs with MoO_x and PEDOT:PSS as the HEL and PTB7: PC_{71}BM as the photoactive layer.

2. Experiment

PTB7 was purchased from One Material, Inc.; PC_{71}BM from Nano. C; and PEDOT:PSS, $(\text{NH}_4)_6\text{Mo}_7\text{O}_{24}\cdot 4\text{H}_2\text{O}$, *o*-dichlorobenzene (ODCB, anhydrous, 99%), isopropanol solvent, and 1, 8-diiodooctane (DIO) from Sigma-Aldrich Co. PFN was purchased from Luminescence Technology Corp. and Al from Alfa Aesar Co.

$(\text{NH}_4)_6\text{Mo}_7\text{O}_{24}\cdot 4\text{H}_2\text{O}$ 0.4 g was added to the blend of isopropanol solvent and deionized water (10 mL, isopropanol: water ratio of 4:1), and used to prepare the solution of $(\text{NH}_4)_6\text{Mo}_7\text{O}_{24}$ because of the better contact ability of isopropanol solvent with a glass substrate. PTB7: PC_{71}BM (1:1.5, wt.%) blends were dissolved in ODCB solution overnight with a PTB7 concentration of 10 mg ml^{-1} . Approximately 3% (1,8-diiodooctane (DIO)/1,2-dichlorobenzene(DCB), v/v) of DIO added to the PTB7: PC_{71}BM blend is helpful in obtaining better photovoltaic results. The PFN solution was dissolved in a methanol solution (0.2 mg ml^{-1} of methanol) in the presence of a small amount of acetic acid ($2\text{ }\mu\text{l ml}^{-1}$). ITO-coated glass ($R = 12\text{--}14\text{ }\Omega\text{ sq}^{-1}$) was cut into $3 \times 3\text{ cm}^2$ chips and used as substrates. Each ITO substrate was ultrasonicated, followed by washing with acetone, isopropyl alcohol, and deionized water for 20 min. Subsequently, the ITO glass substrates were dried under a stream of nitrogen and heated on a hot stage. Finally, an UV-ozone treatment of the ITO glasses was performed for 15 min [22]. The $(\text{NH}_4)_6\text{Mo}_7\text{O}_{24}$ and PEDOT:PSS solutions were spin-coated onto an ITO electrode, which was then thermally treated at 130°C for 15 min. The thickness of the MoO_x layer and PEDOT:PSS layer is 15 and 20 nm, respectively. The blended solution of PTB7: PC_{71}BM was then spin-coated on top of the modified ITO layer and thermally annealed at 130°C in a glove box. The PTB7: PC_{71}BM photoactive layer has a nominal thickness of $\sim 100\text{ nm}$ (with a variation of $\sim 10\text{ nm}$ over the entire film). Next, the PFN solutions were spin-coated on top of PTB7: PC_{71}BM layer in a thickness of 10 nm. Finally, the Al cathode was vacuum-evaporated through a shadow mask to define the active area of the devices ($3 \times 3\text{ mm}^2$) under a pressure of approximately $4 \times 10^{-5}\text{ Pa}$. The control devices were fabricated with a PEDOT:PSS layer acting as the HEL using the spin-coating method with the same device structure. The current-voltage curves were obtained using a standard source measurement unit (Keithley 2400). The *J-V* characteristics of the PSCs were performed in a glove box under illumination at 100 mW cm^{-2} using an AM 1.5G solar

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