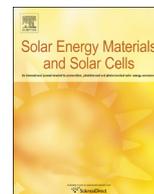




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Contents lists available at ScienceDirect

Solar Energy Materials & Solar Cells

journal homepage: www.elsevier.com/locate/solmat

Enhanced performance of polymer solar cells with PSSA–g–PANI/Graphene oxide composite as hole transport layer



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ARTICLE INFO

Article history:

Received 13 May 2014

Received in revised form

4 August 2014

Accepted 6 August 2014

Available online 3 September 2014

Keywords:

Graphene oxide

PSSA–g–PANI

Hole transport layer

Polymer solar cell

ABSTRACT

A new class of hole transport layer (HTL), which is composed of poly(styrene sulfonic acid) grafted with polyaniline (PSSA–g–PANI) and graphene oxide (GO), was prepared by adding GO into PSSA–g–PANI aqueous solution. The PSSA–g–PANI/GO composites exhibit high optical transparency and high electrical conductivity. When the composite with 2.5 wt% GO loading was used as HTL material for the device based on poly(3-hexylthiophene)/PC₆₁BM blend, the device exhibits an enhanced power conversion efficiency (PCE) of 4.23%, which is 23% higher than the PCE of device with the conventional HTL material, PEDOT:PSS (3.44%). This enhancement of PCE arises mainly from an increase of the short-circuit current density due to higher optical transparency and higher electrical conductivity of PSSA–g–PANI/GO composite. When the PSSA–g–PANI/GO composite is applied as HTL to a low bandgap polymer-based solar cell, PTB7:PC₇₁BM blend, the PCE is also enhanced as compared to the device with PEDOT:PSS, providing us with the possibility to use PSSA–g–PANI/GO as a hole transport layer for various polymer-based solar cells to enhance the photovoltaic performance.

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1. Introduction

Polymer solar cells (PSCs) have many advantages over Si-based solar cells such as low cost, ease of fabrication and the potential for fabrication of flexible and large area solar cells [1]. Remarkable improvements in the performance of PSCs have been achieved by introducing the donor–acceptor bulk heterojunction (BHJ) structure in the active layer for the past decade [2–5]. Recently, low bandgap polymers with broad range absorption of the solar spectrum have been synthesized and used as active layer material to achieve high power conversion efficiency (PCE) [6,7]. Compared to remarkable progress in active layer materials, however, the development of efficient hole transport materials to further enhance the photovoltaic performance of PSCs has been relatively less studied [8,9].

Hole transport layer (HTL) has been used in most of optoelectronic devices, because the introduction of HTL significantly improves both performance and stability of the devices by smoothing the ITO surface, lowering the work function of anode, and enhancing the hole

transport [10,11]. Poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS) has most commonly been used as a HTL material. However, several drawbacks of PEDOT:PSS have been reported such as large particle size of 60–80 nm [12], strong acidic nature of PSS [13], high cost, and low electrical conductivity ($\sim 10^{-3}$ S/cm) [14]. To overcome these problems, graphene oxide (GO) [15–17] and reduced GO [18,19] have recently been used as alternative HTL material to PEDOT:PSS. GO, an oxidized derivative of graphene, is an attractive nanomaterial because of low cost, mass production and solution processability. Li et al. [15] have reported enhanced device performance of PSCs when GO was used as HTL. The enhancement of device performance is mainly due to effective electron blocking by GO and thus suppression of charge recombination. It should be noted that GO with a large band gap (~ 3.6 eV) has slightly lower work function (~ 4.9 eV) than that of the P3HT (5.0 eV) and much higher LUMO level than that of PCBM, which renders efficient hole transport and effective blocking of electron transport to the ITO anode [15,20]. It has also been reported that addition of GO (or rGO) into conductive PEDOT:PSS polymer increases the electrical conductivity, resulting in the improvement of device performance of OLED or PSCs [21–24].

In our previous reports [14,25], a water-soluble and self-doped conducting copolymer based on polyaniline, poly(styrenesulfonic acid)–graft–polyaniline (PSSA–g–PANI), was synthesized and

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utilized as the hole transport material for P3HT:PC₆₁BM-based BHJ solar cells. Due to high optical transparency in the UV–vis region and high conductivity of PSSA–g–PANI, the device with PSSA–g–PANI as HTL exhibited higher PCE than the device with PEDOT:PSS. Hence, taking advantages of both GO and PSSA–g–PANI is expected to further enhance the photovoltaic performance of PSCs.

In this work, we prepared PSSA–g–PANI/GO composite by adding GO into PSSA–g–PANI aqueous solution and used the composite as HTL of PSCs for the purpose to improve the solar cell performance. A series of PSSA–g–PANI/GO composites with different GO loadings were prepared in order to examine the effect of GO composition in the composite on the photovoltaic performance. When the photovoltaic properties of P3HT:PC₆₁BM-based solar cells with the composites as HTL were measured and compared to the reference device with PEDOT:PSS, the device fabricated from the composite with 2.5 wt% GO exhibits an enhanced PCE of 4.23% which is 23% higher than the device with PEDOT:PSS (3.44%).

2. Experimental section

2.1. Materials

A graft copolymer, PSSA–g–PANI, was synthesized according to the method previously reported [25], where the molar ratio of ANI to SSA in PSSA–g–PANI was 0.2. GO was prepared by a modified Hummers method [26]. Regioregular P3HT (55 kDa) was purchased from Rieke Metal Inc. Thieno[3,4-b]thiophene/benzodithiophene (PTB7, 97 kDa) was purchased from 1-material. PC₆₁BM (>99.8+%) and PC₇₁BM (>98+%) were obtained from Nano-C and used as received without further purification. PEDOT:PSS (Clevios P VP AI 4083) was purchased from Heraeus.

2.2. Preparation of PSSA–g–PANI/GO composite

10 mg of GO was dispersed in 5 mL of deionized water via bath-type sonication for 1 h, and centrifuged at 4000 rpm for 10 min to remove aggregated GOs. The GO solution was added into PSSA–g–PANI aqueous solution and stirred for 24 hours. Various PSSA–g–PANI/GO composites were prepared by adding different amounts of GO (1.5, 2.5, 5.0, 7.5, 10.0 wt% with respect to PSSA–g–PANI) into 2 wt% PSSA–g–PANI aqueous solution and stirred for 2 days before use.

2.3. Characterization

The transmittance of HTLs coated onto the bare glass were measured by using an UV–vis spectrophotometer (HP 8452A). The film thickness was determined by using atomic force microscopy. The electrical conductivity was measured by a four-point sheet resistance measurement system (Napson, CRESBOX). Ultraviolet photoelectron spectroscopy (UPS) measurements were performed on an AXIS-NOVA spectrometer (KRATOS Inc.) with He I emission (21.22 eV). The XPS spectra of hole transport materials were obtained using an AXIS-HSi spectrometer (KRATOS).

2.4. Device fabrication and characterization

For fabrication of solar cells with the standard device structure of glass/ITO/HTL/P3HT:PC₆₁BM/Al and glass/ITO/HTL/PTB7:PC₇₁BM/Ca/Al, ITO-coated glass (15 Ω/sq) was first cleaned with acetone and isopropyl alcohol and then dried in air at 120 °C for 2 h. After spin-coating of HTL material on the ITO-coated glass, the device was dried at 120 °C for 30 min in ambient condition. A blend solution of P3HT and PC₆₁BM was then spin-coated at

800 rpm for 40 s, where the blend solution was prepared by dissolving 24 mg of blend (the weight ratio of P3HT:PC₆₁BM=1:0.8) in 1 mL of 1,2-dichlorobenzene and stirring for 12 h in a glove box. Al (100 nm thickness) was thermally evaporated under vacuum lower than 10⁻⁶ Torr on the top of the active layer. The device was thermally annealed at 150 °C for 10 min under nitrogen atmosphere. For fabrication of PTB7:PC₇₁BM-based device, a blend solution of PTB7 and PC₇₁BM was spin-coated at 1000 rpm for 120 s, where the blend solution was prepared by dissolving 25 mg of blend (the weight ratio of PTB7:PC₇₁BM=1:1.5) in 1 mL of a mixture of chlorobenzene:1,8-dioctane (97:3 v/v) and stirring for 12 h in a glove box. Ca (25 nm thickness) was thermally evaporated under vacuum lower than 10⁻⁶ Torr on the top of the active layer followed by Al evaporation (100 nm thickness). The photovoltaic performance was measured under nitrogen atmosphere inside the glove box. The current density–voltage (*J*–*V*) characteristics were measured with a Keithley 4200 source-meter under AM 1.5G (100 mW/cm²) simulated by a Newport–Oriel solar simulator. The light intensity was calibrated using a NREL certified photodiode and light source meter prior to each measurement. The hole-only device was fabricated with the same device configuration as the P3HT:PC₆₁BM-based device except the anode material (Au was used instead of Al). The hole mobility of the device was measured under the identical condition to the condition for optimized photovoltaic cells and calculated from the space-charge limited *J*–*V* curve using the Mott–Gurney law. The external quantum efficiency (EQE) was measured using a lock-in amplifier with a current preamplifier under short circuit current state with illumination of monochromatic light (K3100, McScience Co.).

3. Results and discussion

The chemical structures of PSSA–g–PANI, GO and PSSA–g–PANI/GO composite are represented in Fig. 1. Composites of PSSA–g–PANI/GO are simply prepared by adding GO into PSSA–g–PANI aqueous solution, because both GO and PSSA–g–PANI are soluble in water.

Since light is passed through HTL in front of the active layer, high optical transparency is required for an effective HTL material in PSCs. The transmittances of PEDOT:PSS, PSSA–g–PANI, and PSSA–g–PANI/GO with different GO loadings are shown in Fig. 2, where PANI, PANI/GO(1.5), PANI/GO(2.5), PANI/GO(5.0), PANI/GO(7.5) and PANI/GO(10.0) denote PSSA–g–PANI, PSSA–g–PANI with GO of 0.0, 1.5, 2.5, 5.0, 7.5, and 10.0 wt%, respectively. The PSSA-g-PANI/GO films with 1.5 and 2.5 wt% GO loading show almost the same transmittance as the PSSA-g-PANI film. Although the transmittance of composite films starts to decrease when the GO concentration is further increased above 2.5 wt%, all samples except for PANI/GO(7.5) and PANI/GO(10.0) have higher optical transmittance (over 96%) than PEDOT:PSS in the wavelength range of 450–600 nm, which corresponds to the main absorption range of P3HT as well as other wide band-gap conjugated polymers [27,28].

Another important parameter of HTL for high performance PSCs is its electrical conductivity because increased conductivity of HTL reduces series resistance in the device [14,16]. It has been reported that the short circuit current of PSCs was increased when a PEDOT:PSS modified with sorbitol was used as HTL material, because the electrical conductivity of PEDOT:PSS was increased as its chemical composition was changed by sorbitol [29]. The electrical conductivities of HTL materials used in this study are listed in Table 1. We have already reported that the conductivity of PSSA–g–PANI is higher than that of PEDOT:PSS [14]. To further improve the electrical conductivity of PSSA–g–PANI, GO was

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