



Review

Semiconducting single-walled carbon nanotubes as interfacial modification layers for organic-Si solar cells

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ABSTRACT

A semiconducting single-walled carbon nanotubes (s-SWCNTs) interlayer between poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS) and n-Si was used for high performance organic-Si hybrid photovoltaic (PV) devices. The s-SWCNTs films with different thickness were utilized to investigate the PV effect on PEDOT:PSS/Si device performance. The surface potential of Si substrate with s-SWCNTs was dramatically reduced, which increased the compatibility between Si and PEDOT:PSS. In addition, s-SWCNTs with good semiconducting properties, guaranteed the charge transfer between Si and PEDOT:PSS. Therein, the electrical contact was dramatically improved with addition of s-SWCNTs interlayer, which led to increased fill factor. A power conversion efficiency (PCE) of 12.14% was achieved with an optimized thickness of s-SWCNTs layer. The s-SWCNTs interface layer was fabricated by a simple solution processed method, which was easily coupled with organic-Si solar cells to enhance the PCE.

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1. Introduction

Photovoltaic (PV) industry has been grown rapidly due to the expanding demand for environmental friendly renewable energy resources. Crystalline Si solar cells with the average power conversion efficiency (PCE) of around 20% play a dominant role on the commercial PV market [1]. However, high costs of Si wafers and

complicated manufacturing processes limit large scale application of PV energy. In recent years, organic-Si hybrid solar cells have been extensively studied via simple solution-processed techniques to achieve low cost fabrication process [2]. Conjugated polymers can be easily casted into large-area uniform films by methods such as spin coating, inkjet printing, and spray coating. Up to now, many organic materials have been used to fabricate hybrid organic-Si solar cells. Among them, poly(3,4-ethylenedioxythiophene):polystyrenesulfonate (PEDOT:PSS) attracted wide interests due to its high transparency and conductivity [3]. However, electrical coupling between Si and PEDOT:PSS

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requires to be improved due to its chemical properties difference, where bare Si is usually hydrophobic and PEDOT:PSS is hydrophilic. Hence, it is very important to investigate the organic-Si interface to improve their compatibility.

CNTs display high carrier mobility with tunable electrical and optical properties [4–6]. The conductive CNTs mesh can form a well-defined Schottky junction with Si substrates. High performance PV device based on high conductive CNTs mesh on Si have demonstrated with post-treatment techniques, such as carrier doping of SWNT films by the infiltration of acid, controlling the electronic junctions using ionic liquid electrolyte and coating the Si surface with an antireflection layer. However, active area for CNTs/Si devices is relative small, which is limited by low conductivity of CNTs mesh [7–9]. In addition, CNTs, both multi-walled carbon nanotubes (MWCNTs) and single-walled carbon nanotubes (s-SWCNTs) have been used to function as an interlayer for PV devices due to the excellent charge transfer between organic conjugated polymers and CNTs [10,11]. For example, Somani et al. incorporate double walled carbon nanotubes and metal decorated MWCNTs at the interface between poly(3-octylthiophene) and Si to improve the charge transfer across the organic–inorganic interfaces [12]. Ren et al. demonstrated that the coating of SWCNTs networks on poly(3-hexylthiophene) layers could enhance the charge separation and transportation [13]. Compared to MWCNTs, SWCNTs display many unique advantages for solar cells, such as tunable direct band gaps which could match with wide range of the solar spectrum and excellent charge carrier transport properties. However, SWCNTs are typically grown as bundles of mixed metallic and semiconducting CNTs. Metallic SWCNTs (m-SWCNTs) could function as electrical leads in a nanoscale circuit with quenching the excitons near their vicinity, whereas semiconducting SWCNTs (s-SWCNTs) usually result in nanoscale Schottky-type junctions. Moreover, the mono-dispersed s-SWCNTs exhibit even better characteristics than those of metallic ones.

In this report, s-SWCNTs mesh was inserted between PEDOT:PSS and n-Si by a facile solution processed method to improve contact between them. The s-SWCNTs layer improved the compatibility between Si and PEDOT:PSS. Device structure was Ag/PEDOT:PSS/s-SWCNTs/n-Si/Al, where all photocurrents were generated from Si light harvesting. An induced strong inversion layer was formed in Si once PEDOT:PSS layer was deposited, which is the primary driving force for charge separation. The s-SWCNTs insertion layer improved charge carrier transport property, resulting in a higher short circuit current (J_{sc}). Finally, a PCE of the hybrid devices with s-SWCNTs interface layers achieved over 12% under air mass (AM) 1.5 illumination condition.

2. Experimental methods

Raw SWCNTs was purchased from Nanointegris. Highly pure s-SWCNTs was prepared according to the reported solution selection method [14]. One-side-polished n-type $0.05\text{--}0.1\Omega/\square$ Si (100) substrates were treated by a two-step chlorination/alkylation process [15,16]. The s-SWCNTs layer on Si wafer was fabricated by “precipitation” method. In brief, Si substrates were immersed into the diluted toluene solution of s-SWCNTs for 5 h. Afterwards, the substrates were took out of the solution and annealed at 120°C for 30 min to remove any residue toluene. PEDOT:PSS solution (CLEVIOS PH1000) mixed with 1 wt% Triton (Sigma Aldrich) and 5 wt% dimethyl sulfoxide was spin-coated onto Si substrates covered with s-SWCNTs. Then the films were annealed in a glovebox at 125°C for 30 min. Silver grid and aluminium electrodes were deposited onto the PEDOT:PSS layer and rear side of Si substrates, respectively. Reference devices without s-SWCNTs layer were also fabricated for comparison.

3. Results and discussion

Raw SWCNTs compose of bundles of both s-SWCNTs and m-SWCNTs. According to the UV–Vis absorption and Raman spectra, the as-prepared s-SWCNTs were in high purity after selective purification process. Absorption spectra of as-purchased SWCNTs before and after purification were shown in Fig. 1(a). Obvious metallic first band (M11) transitions of m-SWCNTs were clearly identified in absorption spectra of mixed SWCNTs without purification process. The purified SWCNTs displayed characteristic absorption peaks of well-resolved second (S22) semiconducting transition peak, which was assigned to s-SWCNTs rather than m-SWCNTs. Meanwhile, M11 peaks disappeared, which indicated a selective sorting of s-SWCNTs. Fig. 1(b) showed Raman spectra of the as-purchased SWCNTs powders and the sorted product of s-SWCNTs. The fluorine peri-xanthenoxanthene conjugated copolymer (PFPXX) dispersed s-SWCNTs exhibited a sharp peak at 1580 cm^{-1} corresponding to the G-band, which further verified the high purity of the s-SWCNTs. According to Raman radial breathing mode (RBM) spectra in the inset of Fig. 1(b), average diameter of the sorted s-SWCNTs was $\sim 1.4\text{ nm}$.

The density of s-SWCNTs on Si need to be optimized in order to achieve efficient charge transport as well as maximized light transparency. Light absorbed by the s-SWCNTs layer did not contribute to photocurrent because generated excitons were rapidly quenched in the vicinity of CNTs. All the photocurrents were generated from harvesting light by Si. Thus the s-SWCNTs layers with various coverage ratio and morphology were fabricated by dipping the Si wafers in s-SWCNTs solutions with either high (h) concentration or low (l) one. Atom force microscope (AFM) images of these two kinds of s-SWCNTs meshes were shown in Fig. 2. The contact between the Coulombic repulsion and the strong intertube Vander Waals attractions among s-SWCNTs facilitated the disaggregation [17]. Regarding to s-SWCNTs mesh fabricated from low concentration solution, half ratio of Si region was covered with s-SWCNTs. Regarding to the film fabricated from the s-SWCNTs high solution, the coverage ratio of s-SWCNTs dramatically increased, resulting in rougher surface morphology.

Device structure of PEDOT:PSS/n-Si hybrid solar cells fabricated with s-SWCNTs interlayer was depicted in Fig. 3(a). Current density–voltage ($J\text{--}V$) characteristics under dark and light illumination were plotted in Fig. 3(b)–(c). And their electrical output characteristics were also summarized in Table 1. Dark current densities of the devices from either s-SWCNTs (h) or s-SWCNTs (l) were slightly reduced compared with reference one. Part of dark current originates from the recombination from front interface. Thus, the reduction in the dark current level could ascribe to suppression of charge carrier recombination at the organic-Si interface. We believed that the PEDOT:PSS/n-Si junction quality was improved by inserting s-SWCNTs layer. The reference device exhibited a PCE of 10.32%, with a J_{sc} of 26.67 mA/cm^2 , a V_{oc} of 0.62 V, and a FF of 0.62. Once the s-SWCNTs (l) interface layer was incorporated, a PCE of 12.14% was achieved, with a J_{sc} of 26.40 mA/cm^2 , a V_{oc} of 0.62 V, and a FF of 0.74. In particular, the FF value was dramatically improved from 0.62 to 0.74. Regarding to the devices with the s-SWCNTs (h), a PCE of 11.20% was obtained with a J_{sc} of 26.18 mA/cm^2 , a V_{oc} of 0.62 V, and a FF of 0.69. The reduction in PCE for the devices with the s-SWCNTs (h) was ascribed to the aggregated s-SWCNTs clusters on Si surface after dipping in concentrated s-SWCNTs solution, as shown in Fig. 2(a). Rougher surface morphology of s-SWCNTs covered Si substrates led to poor PEDOT:PSS film quality. In addition, individual s-SWCNTs aggregation in large clusters might protrude out of the PEDOT:PSS layer and contact with the metal cathodes directly, which increased detrimental shunting pathways. As a comparison, the electrical output characteristics of the device

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