



Efficiency evaluation in solar cell by chemically processed hierarchically stacked debundled pristine carbon nanotubes



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ABSTRACT

Pristine SWCNT and MWCNT were individualized and debundled in an N-N dimethyl tetraformamide solvent by a combination of ultrasonication and centrifugation. The wt% (mg) of the pristine CNTs loading were optimized with respect to quantity of solvent (ml). Choice of solvent, ultracentrifugation speed and ultrasonication time were essential parameters for obtaining good individualization of pristine CNTs. By using such CNTs, a thin nanotubes layer (~15 nm) were inserted at the different hierarchical positions into bulk heterojunction solar cell devices made of poly(3-hexylthiophene-2,5-diyl):[6,6]-phenyl-C61-butyrac acid methyl ester with weight ratio of 1:1. By inserting such a pristine SWCNTs between PEDOT:PSS and P3HT:PCBM layers, an increase in the solar cell efficiency from 1.51% (J_{sc} of 6.68 mA/cm², V_{oc} of 0.60, FF of 37) to 2.65% (J_{sc} of 11 mA/cm², V_{oc} of 0.58, FF of 42) were observed. However, functionalized CNTs shows a degrading efficiency of 0.25% which can be attributed to degradation of corrugated tubular surface side walls leading to potential loss of their optoelectronic properties. The enhanced efficiency of devices with pristine SWCNTs can be conjectured to better opto-electrical properties, enhanced transition, improved local structure and the undamaged tubes. The microstructures of the heterojunction active layer were examined by using AFM, TEM, UV-Vis spectra, IV curve and EQE techniques.

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1. Introduction

The steeping energy costs with concerns over the global climate change and demand for clean energy technologies has spurred great deal of interest in organic photovoltaics (OPV) in order to capture the abundant solar energy to generate affordable cheaper electricity resources. The conjugated polymers are one excellent and viable candidates due to their potential low-cost, flexibility, light-weight, colorful and can be solution processed and printed like inks [1–3]. Till date, bulk-heterojunction is the dominant concept which is based on poly(3-hexylthiophene-2,5-diyl):[6,6]-phenyl-C₆₁-butyrac acid methyl ester (P3HT:PCBM) interface, where the three most essential criterion of solar cells such as efficient exciton diffusion, exciton dissociation and charge collection after absorption of solar irradiation are better utilized at the respective electrodes [4–6].

Significant effort has been made towards the interface engineering at the electrodes to improve the power conversion efficiencies (J_{sc} , V_{oc} , and FF) [7,8] by simply blending electron donors with

electron acceptor in organic solvents, self-assembling interpenetrating network on the nanometer length scale obtained by various coating techniques such as doctor blading, [9] slot-die coating, [10] gravure, [11] and inkjet printings [12]. However spin coating is still the dominant practice at the laboratory scale fabrication of OPVs [13]. Apart from spin casting, many effective processing has been developed to optimize the interpenetrating network such as thermal annealing, [14] mixing solvents, [15] additives [16] and hierarchical placement of p-type and n-type buffer layer at various hierarchical positions. One interesting strategy could be the introduction of one-dimensional (1D) carbon nanotubes (SWCNT and MWCNT) in the polymer films to address the series resistance without compromising the shunt resistance of the cells [17]. CNTs are generally inert and have high transmittance in the visible region which hold dual promises as exciton dissociating centers and exciton conductive agents, [18] possess high mobility (~100,000 cm²V⁻¹s⁻¹) [19] and superb electrical, [20] and high thermal conductivities [21]. A combination of CNTs with polymers offer not only to reinforce polymer films but also have the possibility to introduce new electronic properties. Till date, two familiar methods to incorporate CNTs in a BHJ device configuration have been practiced: either as a layered assembly at desired locations or blending with the organic species [22,23]. The success reported with the latter approach depends on chemical

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functionalization, individualization, and length-shortening of the tubes. However, chemical functionalization has been done via concentrated acid treatment, which tends to make these tubes hydrophilic and incompatible for blending with photoactive polymers, [24] besides creating many defects on the CNTs sidewalls and thus have degrading effect on optoelectronic properties. Moreover, additive-assisted non-covalent approaches can facilitate stable suspensions, but again presence of unwanted additives tend to affect optoelectronic behavior [25]. So, it has become increasingly intriguing to stack and utilize the functionalize CNTs at different locations in the BHJ due to its processing bottlenecks [26]. Here in we report a reproducible, scalable simple room temperature laboratory method of incorporating pristine SWNTs and MWCNTs between the PEDOT:PSS and poly(3-hexylthiophene-2,5-diyl):[6,6]-phenyl-C61-butyric acid methyl ester (active) layer in BHJ solar cells, in our study we compared devices made of both pristine CNTs and well known acid treated (functionalized) methods. The pristine CNTs were first individualized and debundled in the optimum N-N-dimethyl tetraformamide solvent and then incorporated as an interpenetrating stack by spin casting. The reason for choosing an amide solvent is based on their suitability for individualization of CNTs [27]. The cells were fabricated by precisely placing CNTs at different hierarchical levels in the device structure. Our study shows that without functionalization (known acid treatment method); CNTs can be successfully spun casted as an interpenetrating layer for efficiency evaluation. Consequently, the BHJ device possesses the increased J_{sc} by minimizing series resistance for maximizing charge collection efficiency regardless of the pristine CNTs applied.

2. Experimental

Reagent grade chemicals [6,6]-Phenyl C₆₁ butyric acid methyl ester (PCBM), Poly (3-hexylthiophene-2, 5-diyl) (P3HT) purchased from Aldrich company. Carbon nanotubes were purchased from Hanhwa Nanotech Korea, and N-N-dimethyl tetraformamide (Junsei Chemicals). Bulk heterojunction polymer solar cell were prepared as follows. The ITO (resistivity 15 V/sq) glass was masked, etched and cleaned with the detergent, then ultrasonicated in acetone and after drying it, cleaned further with O₂ plasma (40 W, 7.5 × 10⁻⁵ Pa) for 15 min. Highly conducting poly(3,4-ethylenedioxythiophene): polystyrene sulphonic acid (PEDOT:PSS) purchased from Baytron (H.C.Stark), was then spin casted (3000 rpm for 40s) over the ITO layer after passing through a 0.80 μm filter. After that the substrate was baked at 120 °C for 10 minutes under vacuum. SWCNT and MWCNT were purchased which contains 30wt% and 90wt% purity, respectively. Firstly the SWCN/MWCNT was dispersed in N-N dimethyl tetraformamide (2 mg/20 ml) respectively, and then sonicated for 1 h, after that it was ultracentrifuge at 1800 g for 30 min. The supernatant was used for spin coating at 3000 rpm/20s at PEDOT:PSS layer (~15 nm) in air. Chloroform used as solvent for active layer application in BHJ. In the 1.0 ml of solvent P3HT (10 mg) and PCBM (10 mg) were added in a 4 ml vial in the glove box then sonicated for 15 min to dissolves it homogeneously, further the solution was magnetically stirred for 24 h (aging). After that active layer was spin casted over the CNTs layer (1000 rpm for 30s) then dried at room temperature. Then with the use of cotton bud we removed the P3HT:PCBM spill over at the PN electrodes. After this procedure film morphology was observed with LASER microscope. To make the cell more simple without utilization of thin LiF and CaF layer an Al (70 nm) electrode was deposited by thermal evaporation in vacuum at about 2 × 10⁻³ torr. Then post annealing was carried out in vacuum at 75 °C for 15 min. For the oxidation the commercially purchased CNTs are refluxed in concentrated H₂SO₄:HNO₃ for 1 h

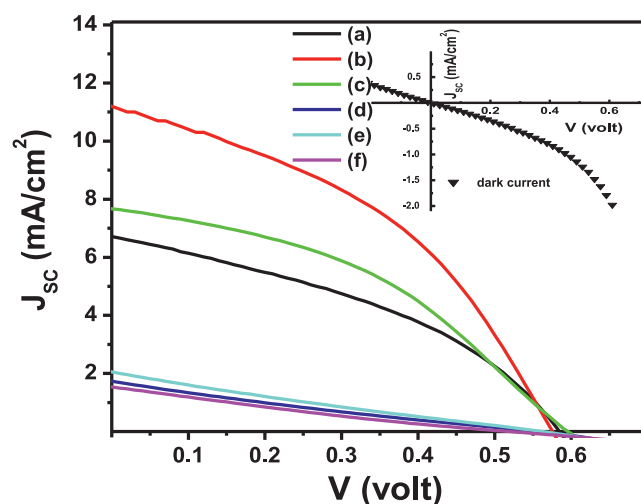


Fig. 1. J-V characteristics of the prepared BHJ solar cells devices (a) ITO/PEDOT:PSS/P3HT:PCBM/Al (b) ITO/PEDOT:PSS/SWCNT/P3HT:PCBM/Al (c) ITO/PEDOT:PSS/MWCNT/P3HT:PCBM/Al (d) ITO/PEDOT:PSS/OxiSWCNT/P3HT:PCBM/Al (e) ITO/PEDOT:PSS/OxiMWCNT/P3HT:PCBM/Al (f) ITO/PEDOT:PSS/P3HT:PCBM/SWCNT/Al; **Inset:** Dark characteristics of a representative devices.

and washed with copious amount of distill water until neutral pH, thus denoted as OxiCNTs.

Devices with different layer compositions were fabricated, which had the structure as follows:

- ITO/PEDOT:PSS/P3HT:PCBM/Al.
- ITO/PEDOT:PSS/SWCNT/P3HT:PCBM/Al.
- ITO/PEDOT:PSS/MWCNT/P3HT:PCBM/Al.
- ITO/PEDOT:PSS/OxiSWCNT/P3HT:PCBM/Al.
- ITO/PEDOT:PSS/OxiMWCNT/P3HT:PCBM/Al.
- ITO/PEDOT:PSS/P3HT:PCBM/SWCNT/Al

Hereafter, (a)–(f) are used for expressing the device structures.

Ultraviolet visible (UV-Vis) absorption spectra were obtained by using a (Hitachi U-4100 UV/Vis/NIR) spectrometer. The morphologies of the CNTs were examined using a Hitachi H-9500 transmission electron microscope (TEM). Atomic force microscopy (AFM) was measured in tapping mode in air by Nano Scope IIIa (Digital Instruments). Current-voltage (J-V) of the solar cells were measured in air with a Bunkoh-Keiki CEP-015 photovoltaic measurement system under a simulated AM 1.5G irradiation (100 mW/cm²). The light intensity was calibrated to 100 mW/cm² using a calibrated standard silicon solar cell and KG5 filter. An array device each having an active area of 0.02 cm², was fabricated on the electrochemically grown substrates and tested under ambient conditions.

3. Result and discussion

Fig. 1 shows the J-V characteristics of the fabricated devices (a)–(f) in a bulk-heterojunction concept. Their detailed solar cell properties are shown in the Table 1. It was observed that when

Table 1
Showing details of J-V characteristic of the fabricated BHJ solar cells.

BHJ Devices	J_{sc} (mA/cm ²)	V_{oc} (volt)	FF (%)	Efficiency% (η)
(a)	6.68	0.60	37	1.51
(b)	11.19	0.58	42	2.65
(c)	7.80	0.60	43	1.82
(d)	2.1	0.56	22	0.25
(e)	1.73	0.55	21	0.21
(f)	1.71	0.53	19	0.17

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