

Effect of hybrid carbon nanotubes–bimetallic composite particles on the performance of polymer solar cells

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ABSTRACT

Hybrid carbon nanotubes–bimetallic composite nanoparticles with sea urchin-like structures (SU-CNTs) were introduced to bulk heterojunction polymer–fullerene solar cells to improve their performance. The SU-CNTs were composed of multi-walled CNTs, which were grown radially over the entire surface of the bimetallic nanoparticles composed of Ni and Al. SU-CNTs with a precisely controlled length of $\sim 200 \pm 40$ nm were dispersed homogeneously in a polymer active layer. Compared with a pristine device (i.e., without SU-CNTs), the SU-CNTs-doped organic photovoltaic (OPV) cells showed an improved short-circuit current density and power conversion efficiency from 7.5 to 9.5 mA/cm² and $2.1 \pm 0.1\%$ to $2.2 \pm 0.2\%$ (max. 2.5%), respectively. The specially designed SU-CNTs have strong potential as an effective exciton dissociation medium in the polymer active layer to enhance the performance of organic solar cells.

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1. Introduction

Solar cell is a potential renewable energy system that converts sunlight to electrical power directly. Among the various types of solar cells, the industrial applications of inorganic solar cells (e.g., silicon-based solar cells) to replace non-renewable energy sources are still limited because of their high fabrication cost. Unlike inorganic solar cells, organic photovoltaic (OPV) cells have attracted considerable attention due to the advantages of flexible, light-weight, and low-cost applications of solar energy conversion [1–7]. Tang [1] reported the development of an OPV with a power conversion efficiency (PCE) of $\sim 1\%$ based on a single donor–acceptor heterojunction. Since then, the PCE has been improved steadily to approximately 5% by many research groups [8–11]. In addition, according to the results of a recent computer simulation, the PCE of OPV cells is predicted to reach 10% by optimizing the donor/acceptor energy levels [12].

The PCE of bulk-heterojunction (BHJ) polymer photovoltaic cells is associated with the open-circuit voltage (V_{oc}), short-circuit current (J_{sc}), and fill factor (FF) delivered by these devices under sunlight illumination. Recently, it was demonstrated that the V_{oc}

of BHJ devices is related to the highest occupied molecular orbital (HOMO) level of the donor [12]. New polymers with a high oxidation potential are needed to increase the V_{oc} . On the other hand, the current density is related directly to the exciton dissociation rate and charge mobility in semiconducting materials, in which the exciton dissociation between donor and acceptor materials should be maximized and sufficient charge collection should be made at the electrodes.

With an aim of improving the performance of OPVs, carbon nanotubes (CNTs) have been introduced recently to OPV devices [13]. The CNTs are used as electrodes [14–16], layered at a desired location [17] or blended with a polymer [13,18–20] as BHJ devices. This is because CNTs are not only optically transparent, flexible and environmentally resistant, they also have strong potentials as exciton dissociating centers and conducting agents with high carrier mobility [18]. However, the applications and characterization of CNTs in these OPV devices have been limited due to the difficulties in incorporating CNTs in polymers. In general, there are two ways of incorporating CNTs in a BHJ device configuration: (i) layer-by-layer assembly of layer-by-layer of CNTs/polymer at the desired locations or (ii) fabrication of a hybrid CNTs–organic species blend. The success of the latter approach is strongly dependent on appropriate chemical functionalization of CNTs, length-shortening of CNTs, and optimization of their concentration in organic composites. Among the range of chemical functionalization methods available, the acid treatment

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of CNTs results in surface modification from hydrophobic to hydrophilic properties so that they can be blended easily with a solution of photoactive polymers [20]. However, OPV devices employing polymer/CNTs composite thin films as an active layer have not achieved a satisfactory PCE so far. This is because CNTs are poorly dispersed in the polymer matrix, and the length of CNTs ranges from a hundred nm to several μm , which makes them larger than the overall thickness of the photoactive layer so that electrical short circuit occurs.

This paper describes a new method for fabricating OPV with an active layer of a hybrid CNT-doped BHJ polymer-composite thin film, where a homogeneous dispersion of CNTs in a polymer matrix was made and the length of the CNTs was controlled precisely. In this approach, the hybrid CNT–bimetallic composite nanoparticles (hereafter called sea urchin-like CNTs [SU-CNTs]) were made by a combination of conventional spray pyrolysis and thermal chemical vapor deposition (CVD). The SU-CNTs-doped OPV cells without chemical functionalization were also characterized. The resulting SU-CNTs were composed of multi-walled CNTs with a diameter of $\sim 10 \pm 2$ nm and a length of $\sim 200 \pm 40$ nm, which were formed over the entire surface of the bimetallic nanoparticles (Ni/Al) with an average diameter of ~ 150 nm. The OPVs composed of P3HT:PCBM heterojunction solar cells incorporating SU-CNTs showed improved performance compared to the pristine OPV cells.

2. Experimental details

The SU-CNTs were produced by a combination of spray pyrolysis and thermal chemical vapor deposition (CVD) in the gas phase, as described in detail elsewhere [21]. Briefly, aluminum nitrate ($\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$, Sigma Aldrich) and nickel nitrate ($\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$, Sigma Aldrich) were first dissolved in deionized water at a 1:1 molar ratio with a total concentration of ~ 3 wt%. An ultrasonic nebulizer was used to aerosolize the mixed metal nitrate precursor solution. The metal nitrate containing aerosol droplets were then transported rapidly using a controlled amount of nitrogen carrier gas ($< \sim 5$ l pm). After the droplets passed through the subsequent silica-gel dryer, the evaporation-induced metal nitrate solid nanoparticles were formed continuously. The resulting metal nitrate nanoparticles were then mixed with hydrogen gas at a flow rate of ~ 100 sccm in a quartz tube enclosed by an electric furnace and heated to ~ 1000 °C so that the metal nitrate nanoparticles were decomposed thermally into pure bimetallic nanoparticles containing both catalytic nickel (Ni) sites in a noncatalytic aluminum (Al) matrix. These Ni–Al bimetallic nanoparticles formed were then introduced into a quartz reactor enclosed by a second vertically mounted tube furnace, where they were mixed with acetylene (C_2H_2) gas at a flow rate of ~ 15 sccm and heated to ~ 750 °C to grow the CNTs on the surface of the catalytic Ni sites in the bimetallic nanoparticles. In order to obtain short CNTs on the bimetallic particles, the residence time of the seeded bimetallic nanoparticles in the thermal CVD reactor was minimized at ~ 10 s by supplying nitrogen gas with a flow rate of ~ 5 l pm. The resulting hybrid CNT–bimetallic composite particles were finally collected on a membrane filter with a pore size of ~ 200 nm.

The OPV device fabricated on an ITO-coated glass substrate was first cleaned in an ultrasonic bath containing acetone and isopropyl alcohol, and then dried with blowing nitrogen gas. A buffer layer of poly(3,4-ethylenedioxyethiophene) (PEDOT-PSS, Baytron P):isopropyl alcohol (IPA) (PEDOT-PSS:IPA=1:2) was made using a spin coater after passing through a 0.45 μm filter with a thickness of approximately 40 nm. The coated PEDOT-PSS film was dried at 150 °C for 1 min on a hot plate

inside a glove box. The photoactive layer was then deposited by spin-coating a mixture of a SU-CNTs–dispersed P3HT:PCBM(1:1) solution with a thickness of 270 and 560 nm, respectively. A 150 -nm-thick aluminum cathode was deposited by thermal evaporation through a shadow mask, which defines a device area of 0.09 cm^2 . Finally, the OPV device was thermally annealed at 150 °C for 20 min. The current density–voltage (J – V) characteristics of the OPV devices were measured under AM1.5 simulated illumination with an intensity of 100 mW/cm^2 (Pecell Technologies Inc., PEC-L11 model) [22]. The intensity of sunlight illumination was calibrated using a standard Si photodiode detector with a KG-5 filter. The J – V curves were recorded automatically with a Keithley SMU 2400 source meter by illuminating the OPV cells.

3. Results and discussion

On the basis of the combination of conventional spray pyrolysis and thermal CVD, CNTs were radially grown on the seeded Ni–Al bimetallic particles, which resembled the “sea urchin” structure. The major advantage of hybrid CNT–bimetallic composite particles (i.e., SU-CNTs) is relatively easy to control the length of CNTs with uniform diameter by changing the residence time of bimetallic particles in the thermal CVD reactor. Another advantage is that we can easily make stable suspension of colloidal particles without additional surfactant due to relatively high specific weight of metallic core structure. As shown in Fig. 1, SEM and TEM images confirmed that multi-walled CNTs (i.e., ~ 15 walls) with a diameter of $\sim 10 \pm 2$ nm and a length of $\sim 200 \pm 40$ nm were formed over the entire surface of bimetallic nanoparticles with ~ 150 nm in diameter (see Fig. 1c).

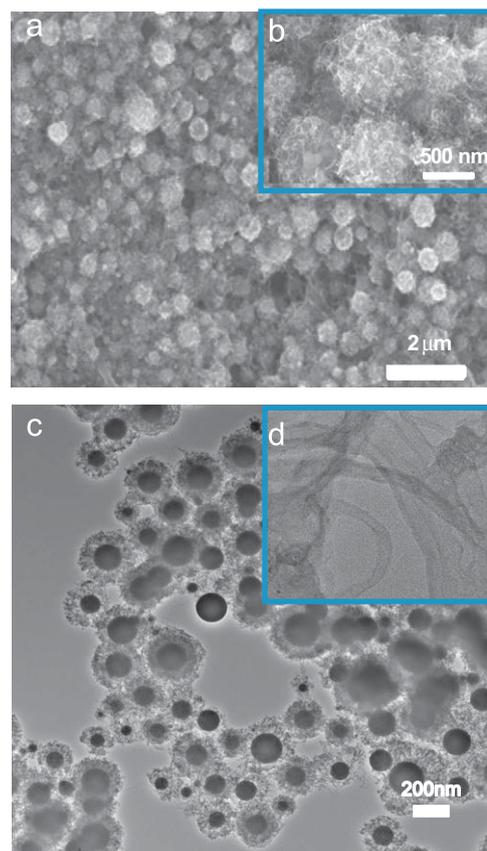


Fig. 1. (a) SEM image of sea urchin-like CNTs ((b) inset: HRSEM image of SU-CNTs) and (c) TEM image of SU-CNTs ((d) inset: HRTEM image of SU-CNTs with ~ 15 multi-walls).

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