



Preparation and employment of carbon nanodots to improve electron extraction capacity of polyethylenimine interfacial layer for polymer solar cells



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ABSTRACT

The performance of polymer solar cells is substantially enhanced by introducing carbon nanodots as additives in polyethylenimine buffer layer. The most pronounced effect is observed in one type of device with the average power conversion efficiencies increased from 5.78% to 7.56% after the addition of carbon nanodots at an optimal concentration in the interfacial layer, which is mainly attributed to the enhanced light trapping and electron transfer in the devices. Besides the light-harvesting and electron transport capacity improvement, the addition of carbon nanodots can also increase exciton generation and dissociation, leading to a high electron mobility. This study demonstrates a facile approach for enhancing the efficiencies of polymer solar cells.

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1. Introduction

As an emerging photovoltaic technology, polymer solar cells (PSCs), which are based on polymer:fullerene composites with a bulk heterojunction (BHJ) structure and solution-processed process, have become a highlight research area in view of their immense potential as promising alternatives for renewable energy generation in recent years [1–3]. One of the most concerned and critical issues of PSCs for commercialization is the power conversion efficiency (PCE) of the devices [4–11]. However, the efficiency of PSCs is not sufficient to meet realistic specifications for widespread commercialization compared with their inorganic counterparts. Recently, many strategies have been employed to improve the PCEs of PSCs based on different principles, such as introduction of buffer layers, additives, noble metal nanoparticles with plasmonic effects, tandem structure, etc. The continuous improvements in device efficiency have been made and PCEs of PSCs have

dramatically increased from about 2.5% to exceeding 10% [11–19].

One limitation of many high-efficiency PSCs is the low electron mobility in the devices, which prohibit the further improvement of the device performance [20–24]. For BHJ PSCs, excitons excited by light absorption need to diffuse at the donor/acceptor interface and dissociate into free charges. Highly efficient PSCs require maximizing charge transport efficiencies and minimizing charge carrier recombination losses. Whereas, there exists a trade-off between the sweep-out of the free charges by the built-in potential and recombination of the free charges due to the low mobility of polymer semiconductors [25,26]. Furthermore, it is still a bottleneck that the optical absorption can be improved by increasing the thickness of active layer, while thicker films usually cause higher charges recombination and larger device resistance, which limits the increase of short-circuit density (J_{sc}) and fill factor (FF). Considerable efforts have been taken to tackle this issue through the design of narrow bandgap donor-conjugated polymers, the invention of novel device structures, the electrode interfacial engineering, and the optimization of processing techniques via proper additives [27–34]. Among them, an effective and convenient approach for pursuing high PCE is to incorporate quantum dots

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(QDs) material into devices, such as directly patterning on the interfaces of anode and cathode, doping into active layers or both [35–38]. As we know, good electrical properties for solar cells such as a low series resistance and a high shunt resistance, which are associated with the contact at photoactive layer/electrodes (superior diode quality) and good bulk conductivity of the device, are responsible for the device efficiency enhancement. Especially, the electrode interface engineering aims at improving the electrical contact between active layer and electrode, which is conducive to efficient charge extraction, so that electrons and holes are selected and transformed from active layer to their respective electrodes [39–42]. Recently, amine-rich polyethylenimine (PEI) has been applied as a hole-blocking layer for PSCs and a high device efficiency has been achieved [43,44]. It is cost-effective, stable in aqueous solutions, environment friendly, and suitable for a solution process at low temperature. However, pristine PEI polymer is insulating with low conductivity and produces a high resistance in a thick film, which usually restricts the photocurrent of PSCs.

In this work, carbon nanodots (Cdots) were incorporated into PEI buffer layer of inverted PSCs based on poly [N-9'-hepta-decanyl-2,7-carbazolealt-5,5-(4',7'-di-2-thienyl-2',1',3'-ben-zothiadiazole)] (PCDTBT):fullerene derivative [6,6]-phenyl-C₇₁-butyric acid methyl ester (PC₇₁BM) to form a high performance cathode interfacial layer for realizing a cell with high efficiency and better stability. Cdots is a good additive to PEI due its good solubility in water, good conductivity, and good film-forming properties. Compared to pristine PEI as the electron transport layer, the PEI modified with Cdots resulted in remarkably increased J_{sc}, FF, and PCE.

2. Experimental section

2.1. Carbon dots preparation and property

The microwave synthesis of Cdots follows procedures given in our previous work [45]. Citric acid (3 g) and urea (6 g) were added to distilled water (20 ml) to form a transparent solution. The solution was then heated in a domestic 650 W microwave oven for 4–5 min, during which the solution changed from being a colorless liquid to a brown and finally dark-brown clustered solid, indicating the formation of Cdots. This solid was then transferred to a vacuum oven and heated at 60 °C for 1 h. A sample of the Cdots was diluted in aqueous solution, which was purified in a centrifuge (10000 r/min, 20 min) to remove large or agglomerated particles.

The morphologies of Cdots were characterized using transmission electron microscopy (TEM). Drops of Cdots dilute aqueous solution were deposited on carbon-coated copper grids for TEM. Morphology characterizations illustrated that Cdots are spherical

and well dispersed (Fig.1(a)). The sizes of Cdots were in the range from 1 to 5 nm. Well-resolved lattice fringes with an interlinear spacing of 0.32 nm of Cdots (Fig.1(b)) are close to the (002) facet of graphitic carbon. The Cdots dilute aqueous solution exhibits excitation-wavelength-dependent PL (Fig.2(a)). The strongest emission was observed centered at 540 nm under 420-nm excitation with PL quantum yield of 18%. The maximum absorption band of Cdots aqueous solution was observed at 420 nm, indicating π - π conjugated structure (Fig.2(b)). The surface functional groups of Cdots were detected using Fourier transform infrared (FT-IR) (Fig.2(c)). Broad absorption bands at 3050–3552 cm⁻¹ are assigned to ν (N–H) and ν (O–H). Absorption bands at 1640–1780 cm⁻¹ are assigned to ν (C=O). The absorption band centered at 1566 cm⁻¹ is assigned to C=N bond. These functional groups improve hydrophilicity and stability of the prepared Cdots in aqueous systems.

2.2. Device fabrication and characterization

The Cdots material was dissolved in deionized water and the concentration was 2 mg/mL. The chemical structure of PEI, PCDTBT, PC₇₁BM are shown in Fig.3(a), and Fig.3(b) are schematic device structure and energy levels. The device structure is glass/indium tin oxide(ITO) (150 nm)/PEI:Cdots(10 nm)/PCDTBT: PC₇₁BM (100 nm)/molybdenum oxide (MoO₃) (4 nm)/silver (Ag) (100 nm). The preparation of devices is described in the following script. Firstly, ITO-coated glass substrates were cleaned in an ultrasonic bath with detergent, acetone, isopropyl alcohol, and deionized water and then dried under a N₂ stream. Then, the substrates were treated using a UV/O₃ photoreactor to eliminate surface contaminants for 10 min. For the electron-transport layer, PEI was dissolved in deionized water (2 mg/mL). Meanwhile, various additive amounts of 0, 0.5 μ L, 1 μ L, 2 μ L, 3 μ L, and 4 μ L Cdots solution were mixed into each 1 mL PEI solution, and corresponding weight ratios (wt.) of Cdots and PEI are 0, 0.5 wt.%, 1.0 wt.%, 2.0 wt.%, 3.0 wt.%, and 4.0 wt.%, respectively. The devices made with different additive concentrations of Cdots were named as Device A, Device B, Device C, Device D, Device E, and Device F. The blend solutions were ultrasonic treated for 30 min, then spin-coated at 4000 rpm on top of the ITO in air. In our experiments, 4000 rpm is the optimal parameter obtained through a large number of trials. The thin film was then heated to 100 °C and kept in glove box for 10 min. For active layer, the 1,2-dichlorobenzene (DCB) solution composed of PCDTBT (7 mg/mL) and PC₇₁BM (28 mg/mL) were spin-cast at 2000 rpm on top of the PEI layer in air subsequently. The fabricated chips with active layer were baked in the atmosphere of argon in the glove box at 70 °C for 20 min. Finally, the cells were fabricated by thermal evaporation of 4 nm MoO₃ and 100 nm Ag as electrode. MoO₃ possessing the highest occupied molecular orbital (HOMO)

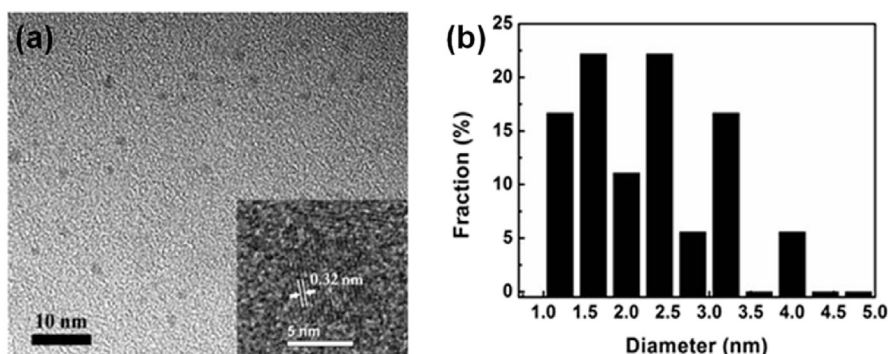


Fig. 1. (a) TEM image of Cdots deposited from water solution on a carbon-coated copper grid, (b) the corresponding size distributions of Cdots in TEM observations.

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