

Quantum dot-nanocarbon based hybrid solar cells with improved long-term performance



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ABSTRACT

In this work, the development of room-temperature solution-processed hybrid solar cells based on CdSe Quantum Dots (QDs) and carbon nanomaterials are reported; multi-walled nanotubes (MWCNT) and reduced graphene oxide (rGO) are incorporated into the photoactive layer of conjugated polymer poly [2,6-(4,4-bis-(2-ethylhexyl)-4H-cyclopenta[2,1-b;3,4-b']dithiophene)-alt-4,7-(2,1,3-benzothiadiazole)], (PCPDTBT) and CdSe QDs. Incorporation of nanocarbon materials helps to improve the long-term efficiency of the solar cells in respect of power conversion efficiency (PCE) and short-circuit current density (J_{sc}) compared to QDs only based devices. For the formation of the hybrid photoactive film hexadecylamine (HDA)/trioctylphosphine oxide (TOPO) capped CdSe QDs were attached to multi-walled carbon nanotubes (MWCNTs) and reduced graphene oxide (rGO) by engineering the interface by introducing thiol functional groups in order to attach CdSe QDs. Initial PCE values of about 2% under AM1.5G illumination have been achieved for these hybrid nanocarbon-CdSe QDs photovoltaic devices. Furthermore, the long term stability of the photovoltaic performance of the devices was investigated and found superior compared to CdSe QDs only based devices. About 84% of the original PCE remained after storage in a glove box for one year without any further encapsulation. It is assumed that the improvement is mainly due to the thiol-functionalization of the nanocarbon interface leading to a strong binding of CdSe QDs resulting in an improved preservation of the nanomorphology of the hybrid film over time.

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1. Introduction

Solution-processed solar cells based on organic–inorganic hybrid materials have been developed as an alternative route towards low-cost fabrication, high efficiency, light-weight and flexible large-area devices [1]. Hybrid solar cells are in principle comparable to that of organic solar cells, the only difference is that inorganic semiconductor QDs are used as electron acceptors instead of fullerene derivatives such as PCBM [2]. The QDs surface ligands [3] and the photovoltaic device architecture [4] are important factors in determining the optoelectronic properties of QDs solar cells. Recent improvement in QDs solar cell power conversion efficiencies up to 5.5% were based on Polymer/PbS_xSe_{1-x} alloyed QDs which showed improved PCEs compared to both conventional PbS or PbSe QDs based devices [5]. However, solar cells based on solution-processed hybrid materials reported so far

generally suffer from a poor device long-term stability in air and over time [6]. Hence, simultaneously achieving the goals of high efficiency, solution-processability and long-term stability in hybrid photovoltaic devices remains a major challenge. Especially the aspect of device stability has been given relatively little consideration and attention by the scientific community so far [7].

The nanoscale morphology of the active layer is a crucial parameter for the device performance of hybrid solar cells [8–10]. The optimization of the nano-phase separation plays a decisive role in increasing the charge carrier mobilities in hybrid solar cells. Hence, the design of continuous charge carrier pathways to the respective electrodes combined with an improved environmental stability should greatly enhance the overall performance of hybrid solar cells. In this context, carbon nanomaterials such as CNTs and graphene, are ideal organic compounds to be combined with inorganic semiconductor QDs. CNTs possess excellent environmental stability and high charge mobilities, which could be utilized to overcome the low electron mobilities in hybrid solar cells [11,12]. There have been already some efforts to incorporate CNTs into solar cells, for example as a hole extraction layer [13,14],

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and as charge transport layer or electrode [12,15]. So far, in general only relatively low PCEs have been reported for CNTs incorporated into the active layer of hybrid solar cells [16]. Nevertheless, by chemical binding of PbS QDs to CNTs, a P3HT:PbS/CNT bulk-heterojunction (BHJ) solar cell with a PCE of 3.03% has been reported [17]. The integration of QDs-graphene nanocomposites into such hybrid solar cells has been recently realized and shows a significant improvement with 4.2% in PCE compared to 2.7% of QDs-only based devices [18,19].

The preparation of nanocarbon-QDs hybrid materials is usually based on the simple addition of QDs to CNTs, utilizing solely Van der Waals interactions between QDs and the surface of the CNTs [20]. Nevertheless, this method tends to result in uncontrolled QDs loading. Using *in-situ* synthesis of semiconductor nanoparticles directly on the surface of CNTs might be considered as an alternatives route. However, this route was found to interfere with the QDs synthesis leading to non-uniform and polydisperse nanocrystals [21,22].

In this work, we apply a simple synthesis method towards thiol-functionalized nanocarbon materials recently reported for the thiolation of reduced graphene oxide (rGO) [18,23]. The successful incorporation of MWCNTs and rGO as an additive into the active

layer of hybrid solar cells based on CdSe QDs are reported. Preservability about 84% of its original PCE after storage in a glove box for one year without any further encapsulation demonstrates a promising potential pathway towards high-performance solution-processed hybrid solar cells with enhanced long-term stability.

2. Experimental

2.1. CdSe QDs synthesis

CdSe QDs were synthesized according to Yuan et al. [24]. First, 2898 mg (12 mmol) of HDA (hexadecylamine, 95%, Merck Schuchardt), 3092 mg (8 mmol) of TOPO (trioctylphosphine oxide, 99%, Sigma-Aldrich), and 444 mg (0.4 mmol) of Cd-stearate were heated up in a nitrogen atmosphere inside a 25 ml three neck flask to 300 °C. After reaching 300 °C, 400 ml (0.4 mmol) of a 1 M solution of selenium in TOP (trioctylphosphine, 97%, ABCR) was rapidly injected. The synthesis was continued under stirring 1000 rpm and stopped after 30 min. QDs with a full width at half maximum (FWHM) value of 24.5 nm, a first excitonic absorption peak at 635 nm, a PL emission maximum at 645 nm, and an average size of 6.3 nm were obtained.

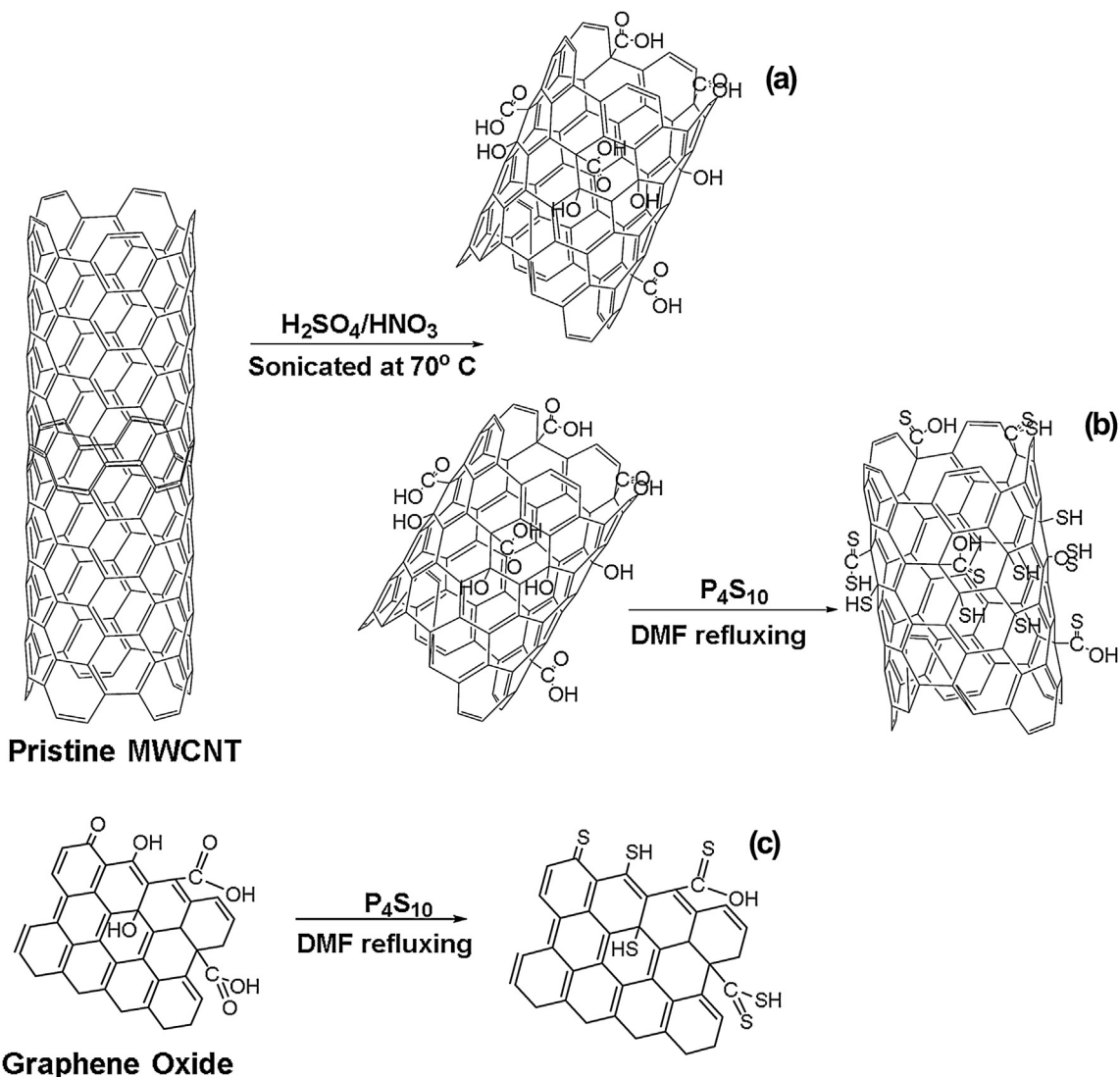


Fig. 1. A schematic illustration of carbon nanomaterials synthesis in this work: (a) NTCNT, (b) TCNT and (c) TrGO. Prior to thiol functionalization, MWCNTs are oxidized and cut by sonication in a mixture of H_2SO_4 (95%) and HNO_3 (65%) with 1:3 volume ratio.

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