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Effect of CdS nanoparticle content on the in-situ polymerization of 3-hexylthiophene-2,5-diyl and the application of P3HT-CdS products in hybrid solar cells

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ABSTRACT

Organic materials are of great interest for photovoltaic applications because of their potential low cost of production that could lead to inexpensive energy harvesting. In this paper, the synthesis and characterization of new nanocomposites of poly(3-hexylthiophene) and CdS nanoparticles (P3HT–CdS) as well as its application in a photovoltaic solar cell is demonstrated. The P3HT–CdS nanocomposites were synthesized by the direct oxidation of 3-hexylthiophene (3HT) monomers with different concentration of CdS nanoparticles. Iron chloride ($FeCl_3$) was used as the oxidizing species for the reaction. The CdS nanoparticles were synthesized by simple solution methods. The P3HT–CdS nanocomposites were analyzed by using UV–vis spectroscopy, Scanning Electron Microscopy (SEM), Fourier Transform Infrared (FT-IR) spectroscopy and Thermogravimetry (TGA). SEM analyses demonstrated homogeneous distribution of the CdS nanoparticles in the P3HT. The interaction of the CdS nanoparticles with the sulfur of the thiophene rings was corroborated by FTIR. Better arrangement in the P3HT chains is also evident from the UV–vis results. Higher CdS content in the P3HT–CdS products reduces the intensity of the π–π* band of the P3HT, suggesting that the polymerization of 3HT monomer could be inhibited for higher concentration of CdS nanoparticles. Hybrid photovoltaic cells based on these nanocomposites were also fabricated and evaluated. The best performance was obtained using as active layer of 3HT/CdS with a weight ratio of 1:0.5. This device showed a photocurrent (J_{sc}) of 1.04 mA/cm^2 and a photovoltage (V_{OC}) of 820 mV. The device was measured in air at room temperature and under 100 mW/cm^2 illumination in a solar simulator.

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1. Introduction

Research for the potential use of semiconducting polymers in photovoltaic solar cells began almost from its discovery in the 1980s [1]. Specifically, solar cells based on soluble conjugated polymers like poly(3-alkylthiophenes) have attracted attention because the use of soluble conjugated polymers might results in inexpensive optoelectronic devices [2]. For solar cell applications, poly(3-hexylthiophene) (P3HT) is one

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of the most promising candidate due to its high absorption coefficient ($\sim 10^5\,\text{cm}^{-1}$) in the visible region of the solar spectrum as well as the high mobility of the photo-generated holes [3]. Polymer–inorganic hybrid solar cells are of particular interest because these cells combine the solution processability of polymers with the high electron mobility of inorganic semiconductors [4]. In particular, the use of semiconductor nanoparticles (NPs) results in improved electronic properties, photoconduction and photoluminescence, as compared with the larger particles [5,6]. Furthermore, bulk heterojunction devices composed of conductive polymers and inorganic semiconductor nanocrystals (NCs) are attractive in photovoltaics mostly for two reasons: a) potential complementary light absorption of the components, and b) better charge separation at the organic–inorganic interface [7]. These bulk heterojunctions have large interfacial area between the two components materials (donor and acceptor), which would benefit the performance of the cells [8]. Bulk hetero–junctions (BHJ) are generally fabricated using an active layer consisting of a conjugated polymer (electron donor) and an inorganic semiconductor NPs as electron acceptor such as CdS, CdTe, CdSe, ZnO [9–18]. Several methods have been reported for the NPs synthesis including solvothermal, sol–gel and co-precipitation, among others [13,19]. Initially these active layers were prepared by mixing the inorganic nanoparticles with conjugated polymers in common solvents [20–22]. Organic capping ligands are necessary due to the poor dispersion of the inorganic nanoparticles into the polymer. Although the incorporation of the surface ligands improves the distribution of the NPs in the conjugated polymer, the insulating nature of such organic ligands results in an electrical barrier for charge transport between the polymer and NPs [23]. To minimize this problem, inorganic nanoparticles have been directly synthesized in the presence of the conjugated polymer matrix without surfactants or ligands. In this case, the sulfur atom in the thiophene ring provides an anchoring site for the nucleation of the NPs [2,24–29].

Semiconducting hybrid systems without ligands can be prepared in-situ by synthesizing the semiconductive polymer in the presence of metal chalcogenides. The hybrid material shows improved interaction between its components and facilitate the charge separation at the donor–aceptor interface [7]. So far, only one report has focused on the synthesis in-situ of semiconducting polymers in the presence of metal chalcogenides, as reported by Ogurtsov et al. [7] in 2011. In that paper the authors report hybrid nanocomposites of poly(3-methylthiophene) and CdSe nanoparticles based on the chemical oxidative polymerization of 3-metylthiophene in the presence of CdSe nanoparticles. Solar cells with the following configuration ITO/PEDOT:PSS/CdSe–P3MT/Al were demonstrated. This demonstrated the possibility to fabricate a solar cell based on a BHJ with a polymer/inorganic hybrid nanocomposites. The active layer contained shells an insoluble photoactive polymer (P3MT) and inorganic semiconductor core of CdSe nanoparticles. No efficiency of the solar cells was reported.

In this paper, the effect of CdS nanoparticle content on the in-situ polymerization of 3-hexylthiophene-2,5-diyl is presented. The physical and chemical properties of the hybrid layers as well as the characteristics of the resulting solar cell were analyzed. The results showed that the P3HT polymer and the CdS nanoparticles formed a bulk heterojunction and that the efficiency of the hybrid solar cell strongly depends on the CdS nanoparticle concentration.

2. Materials and methods

CdS nanoparticles were synthesized by using a precipitation method previously reported [30]. In this method, 0.1 M solution of Na_2S (Fermont) and 0.1 M solution of cadmium acetate $[Cd(OOCCH_3)_2 \cdot 2H_2O$, Alfa Aesar, 99.999 %] were prepared in methanol. The reaction temperature was set at 25 °C (room temperature) and the reaction time to 0.5 h. The cadmium-ion containing solution was slowly added to the sulfide-ion containing solution. This results in a color change from transparent to yellow. This is indicative of the formation of CdS. The obtained CdS precipitates were cleaned by centrifugation 11 times with 50 mL of deionized water each time. One last clean was performed with 50 mL of methanol. All the final products were dried up at room temperature for 48 h.

The synthesis of the nanocomposites of P3HT and CdS nanoparticles (P3HT–CdS) was carried out at 3(hexylthiophene)/CdS (3HT/CdS) weight ratios of 1:3, 1:1.5, 1:0.78, 1:0.5 and 1:0. The P3HT polymerization was performed by the direct oxidation of the 3HT monomer (Aldrich, 99%) using Ferric chloride (Aldrich, $FeCl_3$ 97%) as oxidant at room temperature in an inert atmosphere, as reported in [31]. For the synthesis of the P3HT–CdS composites, the CdS nanoparticles were dispersed in anhydrous $CHCl_3$ in an ultrasonic bath and then added into another anhydrous $CHCl_3$ solution with 0.025 mol of $FeCl_3$ also under ultrasonic conditions. Next, 0.0167 mol of distilled 3HT was dissolved in anhydrous $CHCl_3$ and slowly added into the $FeCl_3$/CdS solution. The reaction mixture was stirred at room temperature for 20 h. The product was then precipitated in methanol, filtered and carefully washed with methanol, hydrochloric acid (10 vol%), acetone, NH_4OH (10 vol%), ethylenediaminetetraacetic acid (EDTA, 1 vol %) and distilled water. The washed P3HT–CdS nanocomposite was then dried. For comparison, P3HT without CdS nanoparticles was also synthesized following the procedure described above.

The P3HT–CdS and P3HT films were deposited on Corning glass substrates by spin-coating with a rotational speed of 3000 rpm from a solution with a concentration of 2 mg/ml of the nanocomposite/chloroform. This solution was stirred under ultrasonic conditions before deposition. UV–vis spectra of the films were obtained in a USB4000 Fiber Optic Spectrometer in a wavelength range of 300–1000 nm. SEM analysis was performed in a Scanning Electron Microscopy (SEM, ZEISS SUPRA-40 Scanning Electron Microscope). FT-IR spectra were recorded on a Bruker Laser ATR Spectrometer. ^{1}H RMN experiment was performed in a Varian Inova 400 Spectrometer, using $CDCl_3$ as solvent and tetramethylsilane as the internal standard. TGA spectra were obtained with a TGA/DSC 1 (Mettler Toledo).

For the solar cells fabrication a 50 nm CdS film window layer (CdS-f) was deposited by chemical bath deposition for two hours at 60 °C on a clean ITO coated glass substrates (Delta Technologies, $R_s = 8–12\,\Omega$ sheet resistance). The composition of the chemical bath solution for the window layer consists of 12.5 ml of cadmium nitrate (1 M), 7.5 ml of

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