

Efficient hybrid polymer/titania solar cells sensitized with carboxylated polymer dye

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ABSTRACT

A facile method to prepare polymer-TiO₂ hybrid nanocomposite based solar cells has been developed. Here a fabrication method to make the nanocrystalline TiO₂ layer from commercial TiO₂ powder using a simple approach is reported. Active layers were prepared in a thickness range from 0.3 to 1.8 μm with this method. We also demonstrate its use to obtain efficient organic-inorganic hybrid nanocomposite solar cells giving photovoltaic current density as high as 4.5 mA/cm² and power conversion efficiency of 0.79%. Commercially available conjugated polymers such as poly[3-(5-carboxypentyl)thiophene-2,5-diyl] (P3HT-COOH) as sensitizer, poly(3-hexylthiophene) (P3HT) as hole conducting layer and conducting poly[3,4-(ethylenedioxy)-thiophene]:poly(styrene sulfonate) (PEDOT:PSS) as charge collection layer have been used. Effect of annealing on the solar cell performance is studied in correlation to the morphology of the nanocomposite as well as absorption in active layers.

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1. Introduction

During the past decade, there has been an intensive research on the fabrication of cost-effective photovoltaics that can act as long-term viable alternatives to the conventional silicon-based solar cells [1–3]. Solar energy conversion based on organic semiconductors is an emerging research field with substantial future prospects and it has attracted great attention due to the advantages of light weight, flexibility and low cost of production with the possibility of fabricating large area devices based on solution processing [4]. Achieving efficient solar energy conversion on a large scale and at low cost is one of the most important challenges for the near future. New materials and fabrication procedures leading to the substantial reduced cost of photovoltaic electricity could help drive a rapid expansion in implementation of photovoltaic technology. The need to improve the light-to-electricity conversion efficiency requires the implementation of such materials and the exploration of new device architectures [5]. Bulk hybrid heterojunctions (BHJ) based on blends of polymers with fullerene derivatives are the best studied devices having highest efficiencies crossing 5%, to date [5–7]. However, the use of fullerenes as electron acceptors has some drawbacks such as segregation of the molecules during aging and relatively poor photostability [8]. Inorganic semiconductors such as TiO₂ [9,10], ZnO [11,12], etc. are promising as alternative electron acceptors and offer good electron transport properties, excellent physical and chemical stability and facile fabrication properties.

Changing the size of these electron acceptors can control the microstructure formation in order to create a large interfacial area when coated with the polymer. Some attempts to increase the charge separation efficiency by increasing the TiO₂/polymer interfacial area have been reported. More than interfacial area, it is the pore diameter and the pore volume in mesoporous metal oxide networks that determines the overall performance of the device and this has been concluded by the comparative study of various TiO₂ pastes prepared by different techniques [13]. Investigation on mesoporous nanocrystalline TiO₂ clearly demonstrates that anatase crystal phase, optimal pore size and a well-connected nanoparticle morphology are essential for the efficient functioning of the solid-state dye-sensitized (SDSC) solar cell [14]. For the best performing TiO₂ electrodes, the synthesis of crystalline TiO₂ and its conversion to screen-printable paste and finally to mesoporous films involve tedious and lengthy procedure running for 2–3 days. Such a long-time procedure is economically unsuitable for industrial production and must be reduced. Attempts to make screen-printable paste based on water and alcohols from a commercially-available crystalline TiO₂ powder induced TiO₂ aggregation. Recently, Ito et al. [15] developed a new short procedure to produce TiO₂ pastes with α-terpineol from commercially available nanocrystals and used it in liquid electrolyte cells. These α-terpineol-based pastes are long-term stable and give very reproducible results. It is of great interest to prepare such pastes and test them in solid state devices.

Morphology and filling with polymer: Low molecular hole conductors exhibit better penetration into the pores of mesoporous TiO₂ film than polymeric counterparts and hence the observed better performance [14,16]. However, it is equally true that conjugated polymeric hole-conductors have better charge

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transport and absorption properties and would lead to inherently more efficient solar cells, which is essential on long-term prospects. It is anticipated that materials with a higher mobility will allow for an increase in efficiency as charges will be transported more quickly to the electrodes and hence reduce the possibility of recombination and higher mobility will allow for a thicker cell to be made, thereby enabling a greater fraction of the incoming sunlight to be absorbed. However such materials have to be processable from solution and should form smooth films. Unfortunately, despite their high mobility, the incomplete filling of the pores with the hole-conducting material gives lower efficiencies of the cells [16]. If the hole-conductor just covers the TiO₂ with a thin layer without filling the voids in the active area, the positive charges have to travel within this thin film, following the shape of the nanoporous TiO₂ surface, which will be much longer than a direct path and this may increase the chances of recombination. A shorter pathway for the charges towards the electrodes can be available as more hole-conductor material is found in the nanopores. Incomplete pore-filling leads to a higher series resistance within the cell. Conjugated polymers such as polythiophenes and poly(phenylene vinylenes) (PPVs) have been considered as promising light sensitizers and/or charge mediators for efficient photovoltaic performance. Polythiophene is an attractive candidate for nanocrystalline TiO₂ photovoltaic (PV) cell due its environment stability and tunable electrochemical properties. Moreover, apart from being advantageous to be used as solid electrolyte, efficient photoinduced charge transfer and well matched energy levels among polythiophenes and nanocrystalline TiO₂ are considered essential requirements to produce efficient solar-to-electric energy conversion in organic molecules sensitized nanocrystalline TiO₂ PV cells [17].

In this paper, we describe the fabrication method to make the nanocrystalline TiO₂ paste by a simple way and we demonstrate its use to obtain efficient organic-inorganic hybrid nanocomposite based solar cells giving photovoltaic current density as high as 4.5 mA/cm² and power conversion efficiency of 0.79%. We used commercially available conjugated polymers such as poly[3-(5-carboxypentyl)thiophene-2,5-diyl] (P3HT-COOH) as sensitizer, poly(3-hexylthiophene) (P3HT) as hole conducting layer and conducting poly[3,4-(ethylenedioxy)-thiophene]:poly(styrene sulfonate) (PEDOT:PSS) as charge collection layer towards Au. Attempts to fabricate solar cells simply by mixing nanocrystalline titania and P3HT-COOH polymer simply did not yield appreciable results in terms of current and efficiency. So we prepared TiO₂ paste and the films made by this paste were sintered and finally P3HT-COOH was chemisorbed on it followed by coating with P3HT. In this way, an efficient hybrid nanocomposite of donor polymer and acceptor TiO₂ was obtained. The fabrication of the mesoporous nanocrystalline TiO₂ paste used for making devices is described in details in the experimental section with some modification in the procedure by Ito et al. [15]. In short, the commercially available nanocrystalline TiO₂ powder (Aldrich: about 15 nm average diameter) was grounded in the mortar and at each step acetic acid followed by water and ethanol were added drop by drop. The TiO₂ dispersions in the mortar were transferred with excess of ethanol to the beaker and stirred. The ultrasonic homogenization was performed using Branson digital sonifier at 60% amplitude. Then α -terpineol was added followed by stirring and sonication. The contents were concentrated by evaporation on rotary. The water added at first can make the surface covered by hydroxides and make strong chemical bonding that make good connection between TiO₂ particles and between a TiO₂ particle and surface. However, it is equally important that the particles should not be coagulated to large aggregates. The aggregates in the matrix can make large shrinkage of the film during sintering process resulting in a peeling-off from the substrate. The acetic

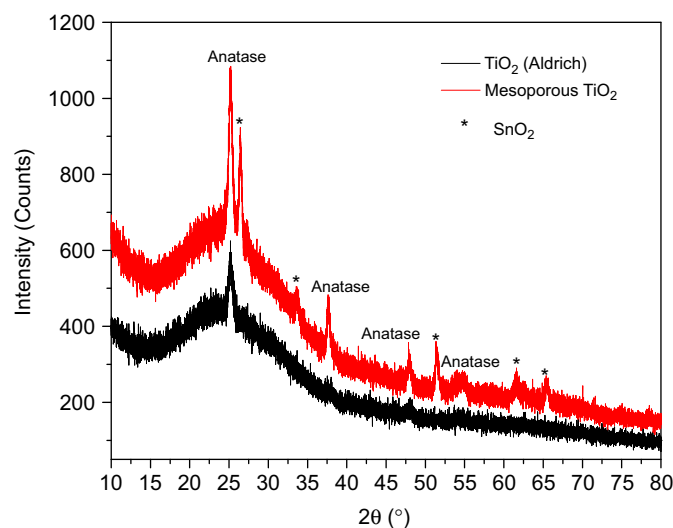


Fig. 1. XRD patterns of TiO₂ nanocrystals (Aldrich) (black) and as-made TiO₂ (red) films deposited on FTO substrates. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

acid adsorbed on the surface of TiO₂ particles prohibits each particle from aggregation [15]. TiO₂ films were prepared by screen-printing on FTO glasses by varying the compositions of as-made TiO₂ paste by diluting with α -Terpineol and the films were finally sintered at 500 °C. X-ray diffraction patterns of the films prepared by as-made TiO₂ and the used commercially available anatase TiO₂ nanocrystals are shown in Fig. 1 for comparison. It is observed that the films made of as-made TiO₂ paste after sintering retained the anatase phase. The diffraction peaks due to SnO₂ in FTO layer are marked by asterisk. The lower intensity of XRD peaks in powder film is due to the fact that the film was very thin and the measurement was done on reflective mode.

2. Results and discussion

2.1. Fabrication and photovoltaic performance of the hybrid polymer/TiO₂ solar cells

Dye sensitization for solid-state dye-sensitized solar cells (SDSC) was carried out by incorporating the carboxylated polymer poly[3-(5-carboxypentyl)thiophene-2,5-diyl] (P3HT-COOH) into the mesoporous nanocrystalline TiO₂ by keeping the substrates in the polymer solution in dimethyl formamide (DMF) for chemisorption for about 18 h and then by rinsing with DMF and drying in vacuum. We used carboxylated P3HT derivative, regioregular poly[3-(5-carboxypentyl)thiophene-2,5-diyl] (P3HT-COOH) (see Scheme 1) with carboxylic functional groups attached to its side-chains as sensitizer. The carboxylate moieties along the polymer side chains provide multiple anchoring sites to the substrate enabling a stable and rugged dye-sensitized TiO₂ film. Krebs [18] recently reported air-stable polymer photovoltaic devices using poly-(3-carboxydithiophene) (P3CT) and ZnO nanoparticles. Polythiophene containing thermally removable solubilizing groups with carboxylic moiety enhanced the interface and the performance of polymer-titania hybrid solar cells [19]. Shankar et al. [10] demonstrated the use of P3HT-COOH polymer in the liquid junction dye-sensitized solar cells as well as solid state solar cells in conjunction with TiO₂ nanotube arrays. Previously, carboxylated polythiophenes were used in regenerative electrochemical photovoltaic cells and correlation of the performances of these cells to the deposition techniques

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