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Influence of loadings of single-walled carbon nanotubes on the performance of ZnO nanorod-based dye-sensitized solar cells

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Abstract

Dye-sensitized solar cells (DSSCs) are a promising low-cost alternative to silicon-based photovoltaic solar cells. Thus, this study was initiated with the aim of combining the unique physical and chemical properties of both SWCNTs and metal oxide nanostructures (such as ZnO-NR) for assembling novel building blocks for solar cells and light energy harvesting devices. DSSCs based on ZnO nanorods/single-walled carbon nanotubes (ZnO-NR/SWCNT) nanocomposite were prepared by the hydrothermal and spin coating method. The photoelectric performances of the DSSCs with various SWCNT loadings up to 1.00 wt% were compared. A gradual increase in the power conversion efficiency (η) was observed up to SWCNT loading of 0.50 wt%, at which the power conversion efficiency of fabricated cells increased to 3.4% compared to 1.97% without SWCNTs. However, further increases in SWCNTs loadings significantly reduced cell performance. Furthermore, the maximum dye absorption intensity was attained at the SWCNT loading of 0.50 wt%. Compared to the DSSC with ZnO-NR electrode, the photocurrent–voltage (J–V) characteristics of the DSSC with 0.50 wt% SWCNTs loading showed an increase in the short-circuit photocurrent (J_{sc}) of 38% leading to an increase in (η) by a factor of approximately 1.7.

Keywords: ZnO nanorods; SWCNT; DSSC; Hydrothermal method

1. Introduction

The heavy reliance on finite conventional fossil-based energy sources over the last several decades along with the associated grave environmental consequences on the one hand, and the sharp increase in their prices on the other, have led to the search for viable alternatives. Solar photovoltaic cells (PV) enjoy the advantage of direct conversion of solar radiation into electricity. However, despite their good efficiency, silicon-based PV cells suffer from shortcomings such as cost and environmental issues [1].

After the breakthrough that came along by the invention of dye-sensitized solar cells (DSSC) by Graetzel in 1991 [2], recent research studies reported an efficiency higher than 11% for this type of solar cells [3]. In addition, due to their advantages like low cost, less toxicity, transparency and flexibility, DSSCs have attracted much attention over the last few years [4]. A typical DSSC consists of a working electrode with wide band gap semiconductor such as TiO_2 or ZnO sensitized using a dye, an electrolyte and a counter electrode. Electron transport and power conversion efficiency of DSSCs depend upon various factors including surface morphology of the working electrode and the dye used. Moreover, cell performance is adversely affected by the low efficiency of transfer of photo-generated charges to the electrodes.

So far, the major part of photoanode DSSC research has mainly focused on TiO_2 nanostructures. However, ZnO semiconductor can be a good alternative as it can exhibit several attractive properties in comparison with TiO_2 semiconductor. Such properties include higher electronic mobility

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for similar band gap energy levels [5], large exciton binding energy which assure efficient emissions and thermal dissociation of excitons, wide direct band gap (3.27 eV), and higher electron mobility that may overcome electron recombination in the TiO_2-based DSSCs [6,7]. Furthermore, several types of ZnO nanostructures with different geometries such as nanoparticles, nanowires, nanorods, or nanobelts, can easily be grown [8–10]. Although efficiencies of ZnO-based DSSCs are still far behind those of TiO_2 [11], ZnO-based DSSCs still constitute a promising candidate as DSSC photoanodes for DSSCs due to their aforementioned photoelectronic properties.

Although nanoparticle-based films can provide a large surface area, the transportation of photogenerated electrons may be hindered or broken off due to the presence of various weak-links and boundaries in the nanoparticle network and thus limiting energy conversion efficiency of DSSCs. Incorporating one-dimensional nanostructures into nanoparticulate films may overcome this problem by providing direct pathways for photoelectron transport [12]. In this regard, carbon nanotubes (CNTs) can provide direct and efficient path for such photo-generated electrons, hence composites of CNTs with metal oxides have been proposed. Sol–gel and electrophoresis methods to synthesize TiO_2-Multiwall CNTs (MWCNTs) nanocomposites have been studied. It was found that the physical and electronic attachment between TiO_2 nanoparticles and the CNT in these studies don't seem to be strong enough to prevent recombination of photo-generated charges. Despite the superior photoelctrochemical properties of TiO_2, it suffers from low electron mobility and the fact that most of its precursors are highly reactive with water which makes it difficult to prepare it in the forms of nanowires and nanotubes in a simple and cost-effective method [13].

Compared with bare nanocrystalline semiconductors, hybrid CNT/metal-oxides nanocomposites like ZnO/CNT-, TiO_2/CNT-, or SnO_2/CNT-based DSSCs represent a promising alternate for the large scale conversion of solar energy into electricity. These hybrid nanocomposites exhibit high surface area and provide better light scattering properties which promote dye loading and light absorption. In such cells, synthetic inorganic dyes, such as ruthenium(II) multi carboxylic complexes, are commonly employed as molecular sensitizers [14]. It has been reported that the nanocrystalline electrode is confronting the maximum photocurrent theoretically achievable with the Ru-based sensitizers (e.g. N719 dye), and the quest for the novel nanostructured photoanode has become prosperous.

The solar energy-to-electricity conversion efficiency of Ru-based sensitizers showed that the number of attached carboxylates on a dye has an influence on the photoelectrochemical properties of the dye-sensitized electrode. For ruthenium complexes, an incident photon-to-current conversion efficiency of 13% at 510 nm was obtained [15,16].

Studies on the efficiency improvements of DSSCs focused on incorporating carbon nanotubes into the working electrodes [17,18], or improving ionic liquid electrolytes [19]. A more common approach utilizes well-aligned carbon nanotubes fabricated by high cost vacuum based techniques such as reactive sputtering or chemical vapor deposition (CVD). An enhancement of the incident photon-to- electron conversion efficiency (IPCE) by a factor of two has been achieved by using TiO_2- decorated SWCNT films [20]. Another study investigated the applicability of single- wall carbon nanotubes (SWCNT) with the functional group deposited on the FTO-glass substrate of the counter electrode for a dye-sensitized solar cell (DSSC). This film of SWCNT/Ag markedly increased the IPCE from 3.9% (conventional DSSC with a thin film of platinum on the FTO-glass substrate of the counter electrode) to 15.3% (DSSC with SWCNT/Ag/acetylacetone) [21]. When assembled on the electrode surface and decorated by metal-oxides nanostructures, SWCNTs offer the possibility of improving charge collection and transporting charge carriers thus improve the performance of the DSSCs.

In this study, ZnO-NR/SWCNT nanocomposites with various SWCNT loadings were synthesized by a simple, cost-effective and scalable hydrothermal process to be used as working electrodes for the assembled DSSCs. Microstructure, photoelectronic and spectroscopic measurements that highlight the influence of the SWCNT loadings on the cells performances were described.

2. Experimental

2.1. Preparation of ZnO-NP/SWCNTs nanocomposites

All chemicals used in this process were analytical grade and were used without any further purification. Zinc acetate dihydrate ($Zn(CH_3COO)_2 \cdot 2H_2O$, Sigma-Aldrich), zinc nitrate hexahydrate ($Zn(NO_3)_2 \cdot 6H_2O$, Sigma-Aldrich) and hexamethylenetetramine (C6H12N4, Sigma-Aldrich) were used as starting materials for the growth of the ZnO nanorods. In a typical experiment, various amounts (0 to 1 wt%) of acid-treated SWNTs ((HipCO) process at CNI, Houston, TX) were dispersed in 100 mL of a 100 mM of zinc acetate dihydrate (98%, Sigma-Aldrich) in a methanol solution and loaded into a 250 mL-vessel. The solution was subjected to magnetic stirring at 25 °C for 30 min. Subsequently, the solution was moved to a water bath ~~and~~ maintained at a temperature of 90 °C for about 2 h with rigorous magnetic stirring. The vessel was slowly cooled to room temperature. Then, 50 μL of Triton-X-100 surfactant was added to facilitate the spreading of the colloid on the ITO substrate. The colloidal gray solution ZnO-NP/SWCNT was sonicated for 30 min and stirred for one hour at room temperature. Before fabricating ZnO-NP/SWCNT thick films with various amounts (0–1 wt%) of SWCNTs, the ITO substrates were ultrasonically cleaned with Triton-X-100, acetone, and water for 30 min and dried in an oven at 80 °C. This colloidal solution was spin coated on ITO surface at 1000 rpm for 5 s and at 3000 rpm for 15 s, then annealed at 350 °C for 30 min. This process was repeated three times to obtain the optimized thickness and homogeneous substrate to be used as a seed for ZnO-NR growth on the ZnO-NP/SWNT film. In the process of ZnO-NP/SWCNT synthesis, SWCNT aggregation could be observed among the particles of each single-walled carbon nanotube. Also, in physical aggregation