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Highly Stable Dye Sensitized Solar Cells with TiO_2/MWCNT Nanocomposite Photoanodes

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ABSTRACT

Ever since the first Dye Sensitized Solar Cells (DSSCs) have been reported in 1991, a considerable research is going on for improving its efficiency. The intensive research has made conversion efficiency to reach 14.3%. Although, fewer efforts have been made towards the study of their stability. This paper deals with the addition of MWCNTs to TiO_2 photoanode and their effect on the stability of the DSSCs. Nanocomposites of MWCNT and TiO_2 nanoparticles have been prepared in two different methods and characterized using FESEM and XRD. An optimum thick photoanodes were prepared using these composites and used in DSSC preparation. The photovoltaic performance of DSSCs was observed for a period of 6 months with an interval of 2 months. A huge decline was observed in the performance of cells made with pristine TiO_2 photoanode whereas the cells with TiO_2/MWCNT nanocomposite photoanode showed stability with a negligible decrease in the efficiency. Cells with nanocomposites prepared in both the methods were able to maintain their short circuit current (J_{SC}) and open circuit voltage (V_{OC}) even after continuous exposure of ambient light for 6 months. The work concludes that the addition of MWCNTs has made DSSCs less susceptible for ambient conditions.

1. Introduction

Dye sensitized solar cell typically consists of a transparent conductive glass substrate coated with mesoporous titanium dioxide layer which is impregnated with light trapping dye molecules, an electrolyte containing iodide/tri-iodide redox couple and a conductive substrate coated with platinum catalyst. DSSCs are the only third generation organic material based solar cells which have significant commercial deployment, such as facility capable of generating ~2000 kWh per year deployed in Switzerland. The scientific community is working towards the integration of DSSCs with electronic products for indoor use, where they can outperform existing technologies such as amorphous silicon [1]. The efficiency achieved by individual lab-scale devices is around 14.3% [2]. For further improvement of its efficiency, different metal oxides (TiO_2, ZnO, Nb_2O_5) with different morphologies (particles, rods, tubes), different composites (TiO_2/CNT, TiO_2/Nb_2O_5), different dyes (Natural, ruthenium based), different electrolytes (water based, acetonitrile based, gel, solid) and different counter electrode (Platinum, graphite, CNT) have been tried by many researchers.

On the other hand, the stability of its performance is one of the key factors which decide the possibility of commercialization. The main factors which contribute to spoiling DSSCs are exposure to light (photo degradation), temperature (thermal degradation) and moisture and leakage of electrolyte [3]. While the leakage issue has been studied by many researchers and tried to fix it by replacing liquid electrolyte with gel and solid one, the contribution of processes like dye desorption, changes in the electron collection at the photoanode is less enlightened. In this aspect, we report the effect of introducing MWCNTs to the TiO_2 photoanode on the stability of the performance. A comparative study has been carried out on the performance and stability of DSSCs made with TiO_2 photoanode and TiO_2/MWCNT composite photoanode. All the photoanodes are prepared with an optimized wt% of MWCNT and optimized film thickness.

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2. Experimental Methods

2.1 Materials

Titanium (IV) isopropoxide (TTIP), tetramethylammonium hydroxide (TMAH), Triton X-100, and other acids and solvents were used as received without any further purification. MWCNTs with a diameter of 20 nm, length 20 µm and purity > 99.9% were the free samples from United Nanotech Innovations Pvt. Ltd. India. Remaining materials like dye- N-719 [$RuL_2(NCS)_2$]: 2 TBA (L=2,2'-bipyridyl-4,4'-dicarboxylic acid; TBA = tetra–n–butyl ammonium), substrate- fluorine doped tin oxide (FTO) with sheet resistance of the conductive coating 7 Ω/sq, Sealant- DuPontTM Bynel® functionalized LLDPE resin with 50 µm thickness, electrolyte- high performance electrolyte (I_3^-/I^-- triiodide/iodide in acetonitrile) catalyst for counter electrode-high conducting platinum (Pt) paste were purchased from Dyesol, Australia.

2.2 Preparation of TiO_2/MWCNT Nanocomposites

Synthesis of mesoporous TiO_2 nanoparticles and the functionalization of MWCNTs were carried out according to our previously reported article [4]. TiO_2/MWCNT nanocomposites having optimized wt% of MWCNTs were prepared in two different ways. In method A, 3 g of nanopowder which is a mixture of TiO_2 nanoparticles and functionalized MWCNTs (f-MWCNTs) in 0.19 wt% were ground with glacial acetic acid (2.25 mL), ethanol (30 mL) and Triton X-100 (300 µL) in a mortar and pestle until a fine paste was formed (A-0.19). In the same way, only TiO_2 nanoparticles were ground to form a fine paste (A-0.00). On the other hand, in method B, a stock solution was prepared by stirring α-terpineol (19 g) and ethyl cellulose (1 g) overnight. The mixture of TiO_2 nanoparticle and f-MWCNTs (3 g) were added to 2.625 g of stock solution and ground for 10 min. Excess of α-terpineol was added and ground until a fine paste of nanocomposite (B-0.19) was obtained. In the same way, the paste of pure TiO_2 nanoparticle (B-0.00) was prepared for comparison purpose.

2.3 Fabrication of Dye Sensitized Solar cells

FTOs were washed ultrasonically with detergent, ethanol, acetone and DI water successively. Pastes were dispersed over FTO by doctor blade technique to generate films with an active area of 0.25 cm² with the help of an adhesive mask. The thickness of the mask is adjusted such way that final thicknesses of the films become approximately equal to 16 µm. After 10 minutes, films were calcined at 500 °C for 30 minutes and cooled to 80

°C. The electrodes were immersed in 0.3 mM N719 dye in the vacuum dye adsorption chamber for 8 hours at dark. On the other hand, counter electrodes were prepared by coating platinum paste on cleaned FTO. Both photoanode and counter electrode were sandwiched with the sealant in between, which also acts as a spacer. The electrolyte is filled with the holes drilled on counter electrode and holes were sealed using bylen.

3. Results and Discussion

Morphological studies of the photoanodes were conducted using field emission scanning electron microscopy (FESEM, Zeiss Σigma) and the images are shown in Fig. 1. MWCNTs used for the experiment had an average diameter of 20 nm. Uniformly distributed TiO_2 nanoparticles with a smooth surface could be observed in case of pristine TiO_2 paste. A large number of pores were seen on the surface of nanocomposites (A-0.19 and B-0.19). It was also seen that TiO_2 nanoparticles were attached to anchoring groups present on the surface of f-MWCNTs. This lead to a structure of high porosity which intern caused higher dye loading and better efficiencies [5-7].

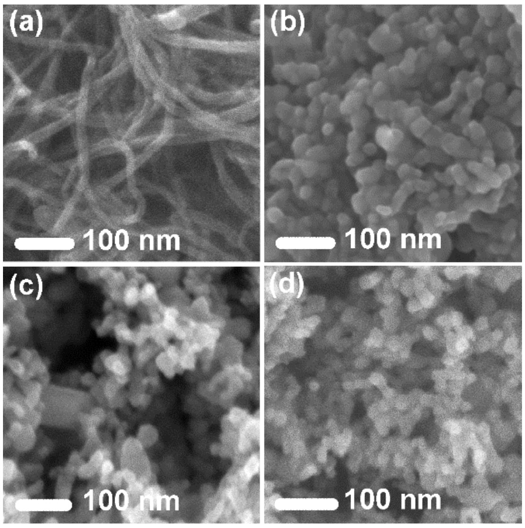

Fig. 1 FESEM images of (a) MWCNTs, (b) A-0.00, (c) A-0.19 and (d) B-0.19 photoanodes

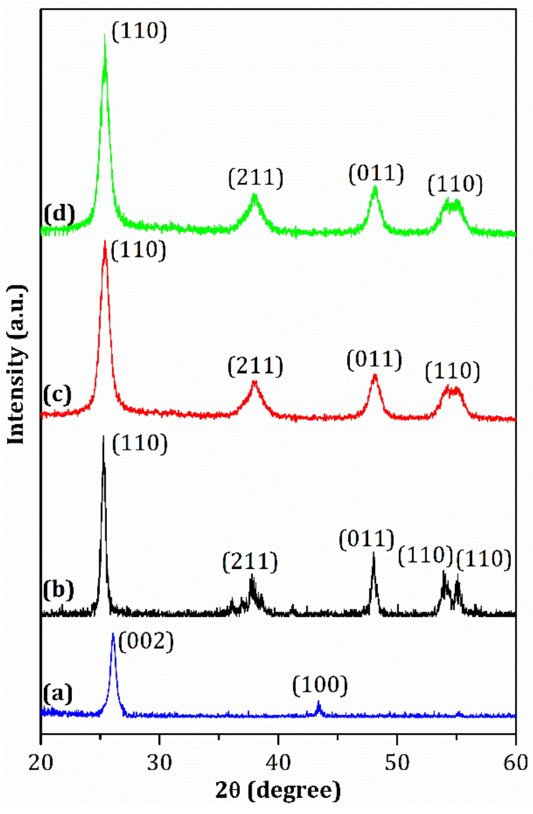

Fig. 2 XRD pattern of (a) MWCNTs, (b) pristine TiO_2, (c) A-0.19 and (d) B-0.19 samples

X-Ray Diffractometer (XRD, Rigaku SmartLab) was used to study structural properties of nanocomposites. Fig. 2 shows the diffraction patterns obtained. Pristine MWCNT exhibited its characteristic peaks [8] at 2θ equal to 26°and 43.4°. According to JCPDS data (84-1286) peaks at 25.29°, 37.84°, 47.95°, 53.91°and 55.06° corresponded to anatase TiO_2 nanoparticles. The crystallinity of TiO_2 was retained even after the addition of MWCNTs. The peaks corresponding to MWCNTs could not be seen in diffractions of both the composites as the crystallinity of TiO_2 was much higher than that of MWCNTs and the main peak of the MWCNT overlapped with the TiO_2 peak corresponding to 2θ value 25.29° making itself invisible [9]. The broadening in the peak represented a decrease in the crystallite size. The average size of TiO_2 crystallite was calculated to be 17.57, 8.51 and 9.67 nm for pristine TiO_2, A-0.19 and B-0.19 respectively from Scherrer's formula ($d = 0.9\lambda / \beta \cos\theta$). This decrease is attributed to the interaction between TiO_2 and the chemical groups such as –OH and –COOH present at the defects of f-MWCNTs. Reduced crystallite size resulted in increased contact points between the TiO_2 particles and between TiO_2 and underlying substrate leading to more efficient charge separation and faster photo-generated electron transfer [10, 11].

The thickness of the generated thinfilm was found using surface profilometer (Mitutoyo, SJ-301). Thickness profiles are as shown in Fig. 3. All the films were uniform with an average thickness of around 16 µm as expected.

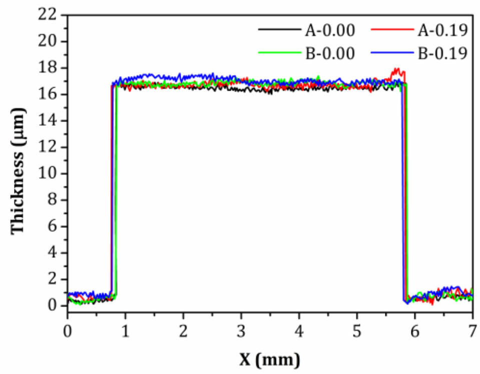

Fig. 3 Thickness profiles of A-0.00, A-0.19, B-0.00, B-0.19 photoanodes

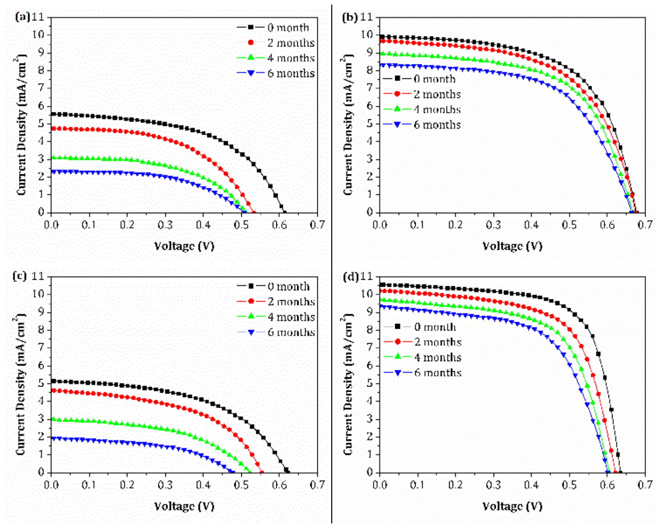

Fig. 4 J-V curves of DSSCs prepared by (a) A-0.00, (b) A-0.19, (c) B-0.00 and (d) B-0.19 photoanodes

The current density as a function of applied voltage was studied using Solar Simulator (Sol3A Class AAA) under the illumination of 100 mW/cm^2 to check the efficiency of the photoanodes and results are shown in Fig. 4. The values are tabulated in Table 1. To determine the stability of the DSSCs all the cells were exposed to ambient light continuously for 6 months and their IV characteristics have been studied with an interval of 2 months. The different parameter as a function of time is shown in Fig. 5. V_{OC} and J_{SC} of DSSCs with TiO_2/MWCNT photoanodes remain almost constant whereas a huge decrease observed for pristine TiO_2 photoanodes, which is a promising factor towards the commercialization of DSSCs. There was a random effect on fill factor. Overall conversion efficiency decreased by 71% and 75% in DSSCs prepared with pristine TiO_2 photoanode for method A and B respectively. But TiO_2/MWCNTs photoanodes helped not only in achieving better efficiency but also in retaining it for a long period of time. Only a small decrease of 21% and 19% was recorded for A-0.19

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and B-0.19 electrodes respectively. This result showed that addition of MWCNTs into TiO_2 photoanode will improve its efficiency as well as stability showing positive hopes towards the commercialization.

Table 1 *J-V* curves of the photoanodes with and without MWCNTs on the day of preparation

Photo-Electrode	Thickness (µm)	Time (months)	V_{OC} (*V*)	J_{SC} (*mA/cm²*)	FF	η(%)
A-0.00	16.51	0	0.614	5.558	0.534	1.82
		2	0.533	4.737	0.528	1.33
		4	0.512	3.083	0.522	0.77
		6	0.507	2.327	0.533	0.53
A-0.19	16.73	0	0.677	9.916	0.604	4.06
		2	0.674	9.676	0.582	3.87
		4	0.668	8.978	0.595	3.53
		6	0.664	8.349	0.589	3.20
B-0.00	16.85	0	0.623	5.150	0.513	1.65
		2	0.550	4.620	0.512	1.29
		4	0.520	3.007	0.492	0.76
		6	0.475	1.925	0.500	0.41
B-0.19	17.07	0	0.635	10.56	0.683	4.80
		2	0.622	10.22	0.638	4.46
		4	0.606	9.725	0.574	4.08
		6	0.601	9.358	0.600	3.88

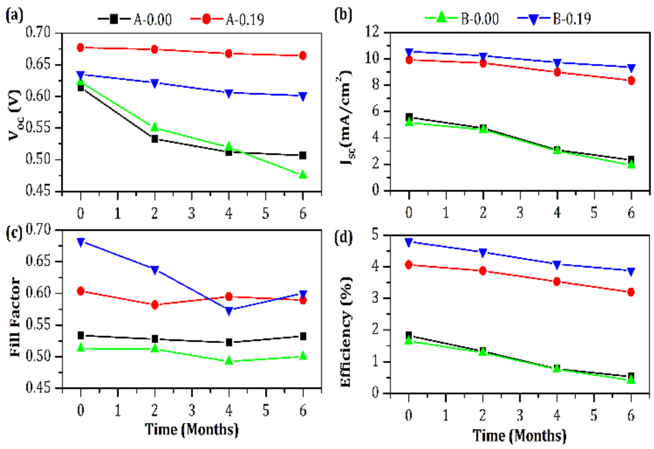

Fig. 5 The time dependence of the parameters of DSSCs with and without MWCNTs in the photoanode, under prolonged ambient light irradiation

4. Conclusion

DSSCs with pristine TiO_2 and TiO_2/MWCNTs photoanodes are successfully prepared and their performance and stability were studied. There was a hike in the performance of the DSSCs with TiO_2/MWCNT photoanode when compared to pristine TiO_2 photoanode. The photoconversion efficiency was increased to 4.06% from 1.82% in the method A and to 4.80% from 1.64% in the method B. Further, they were tested for their stability by exposing them to ambient light and efficiency were recorded for six months with an interval of 2 months. It is observed that efficiency of DSSCs with pristine TiO_2 photoanode decreased by 71% and 75% for method A and B respectively. Whereas DSSCs with MWCNTs in them were able to maintain their performance and their efficiency declined only by 21% and 19% for method A and B respectively.

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