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Enhancement of Photovoltage Generation, Storage Capacity and Energy Conversion Efficiency of Photoelectrochemial Cell of Mixed Dye System: Role of Oxidized Multi-Walled Carbon Nanotubes

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Abstract: In the crisis of fast diminishing fossil fuels, looking for alternative energy sources by utilizing solar energy has the highest research priority and engineered nanoparticles play a very important role here. Using a specially designed photoelectrochemical cell and a mixture of two dyes, Phenosaffranine (PSF) and Azure C (AZC) conjugated with oxidized multi-walled carbon nanotubes (OMWCNTs) we have been able to generate photovoltage of reasonably high magnitude (763 mV). The storage time is also quite high ~ 66 hrs. The photovoltage cycle was reproducible upon further illumination. Energy conversion efficiency (η%) of the cell has been calculated for the system (η% = 4.33). Spectral studies show that with addition of OMWCNTs to a fixed concentration of PSF and AZC solution, the absorbance increases throughout the spectral range. FTIR spectrum reveals that there is no chemical change in the mixed system indicating that only the dyes got adsorbed on the side walls on the OMWCNTs. From fluorescence vspectral study, it has been seen that while fluorescence intensity of PSF quenches with addition of AZC, which does not have any fluorescence of its own, the intensity regains with addition of OMWCNTs. This emphasizes the role of oxidized multi-walled carbon nanotubes in the performance of photoelectrochemical cell of mixed dye system.

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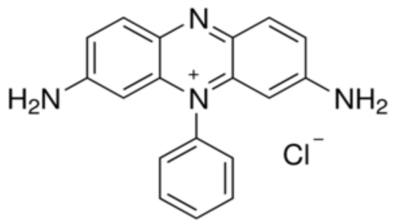
Keywords: mixed dye system; oxidized multi-walled carbon nanotubes; photovoltage; storage capacity; energy conversion efficiency

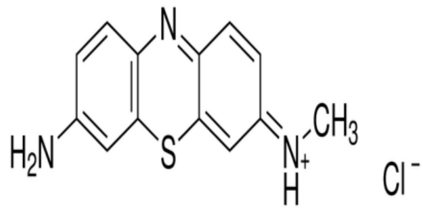
1. Introduction

The depletion of fossil fuels at an alarming rate and their hazardous combustion products pose a serious challenge to the scientists for developing alternative energy sources. In this endeavor utilization of solar energy has become the most promising one. Owing to the limitations of conventional silicon technology based photovoltaics, other cost effective options for harvesting solar energy more efficiently are being explored during the last couple of decades. Use of compound semiconductors caused high optical absorbance and hence good conversion efficiency [1-3]. The invent of nanoparticles, which possess the unique assets of altered chemical and physical properties with size and shape variations, has a strong impact in widening the of possibility in photovoltaic technology [4]. The important factor which limits the performance of conventional solar cells is the mismatch between incident solar spectrum and the spectral absorption properties of the material [5,6]. In the present study we have used photosensitive dyes conjugated with carbon nanotubes as the light energy harvester in photoelectrochemical (PEC) cell. Two dyes having absorption bands in two different spectral regions have been chosen in order to overcome band absorption limits of each dye.

Materials used:

Multiwall Carbon Nanotubes of diameter 2–3 nm were purchased from Arry International, Germany with 60% purity. The dyes Phenosaffranine $[C_{18}H_{15}ClN_3]$ (M.W. 322.79) and Azure C $[C_{13}H_{12}ClN_3S]$ (M.W. 277.77) were purchased from Sigma- Aldrich, India and were used after recrystallization. Cholesterol (E. Merck, Germany) was oxidized and recrystallized from n-octane (E. Merck, Germany). N-decane and iodine were purchased from E. Merck, Germany and before use iodine was purified by resublimation.





Phenosafranine

Azure C

Preparation of Oxidised Multi-wall carbon nanotubes (OMWCNTs):

For oxidation Multi-wall carbon nanotubes (MWCNTs) were at first refluxed with 2M nitric acid at 150ºC for 18 hrs and then sonicated for 6 hrs using an ultrasonic bath sonicator (Model 229; Imeco Ultrasonic, India) in the same acid. Resulting material was collected by filtration and then washed with water and ethanol for several times until the pH of the solution became neutral i.e, 7. After drying the solution in hot air oven at 60ºC for 8 hrs, we got oxidized MWCNTs (OMWCNTs) and those were well dispersed in water owing to presence of hydrophilic groups, such as carboxyl (-COOH), hydroxyl (-OH) and carbonyl groups (>C=O) along their side walls [7].

Preparation of mixed dye-OMWCNTs system:

The OMWCNTs were dispersed in water (concentration 1 mg mL^{-1}) and concentration of aqueous stock solution of both dyes was $1x10^{-4}$ M. The required concentrations of dye solutions for photovoltage (PV) studies were prepared after further dilution of the stock solutions. Individual dye solution was then mixed with OMWCNTs solution and sonicated in an ultrasonic bath for 4 hrs at room temperature and then different dye-OMWCNTs solutions were mixed together in a desired specific ratio by diffusion method.

Absorption spectra of dye-OMWCNT conjugates were recorded in a PERKIN ELMER Lambda 25 UV/VIS Spectrometer (Shelton, CT064844794).

Details of the experimental set-up:.

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Table 1. The characteristics of the PEC cell.

Different parameters	Barrier: Planar lipid membrane	
	Sample Used	
	PSF & AZC	PSF & AZC adsorbed CNTs
Open circuit voltage (Voc) in mV	254	763
Short circuit current in µA	3.9	54.2
Time taken for Voc in hrs	0.8	1.5
Decay time	5.7	66.5
Storage duration in hrs	5	66
Fill factor	0.09	0.38
Energy conversion efficiency (η%)	0.005	4.33

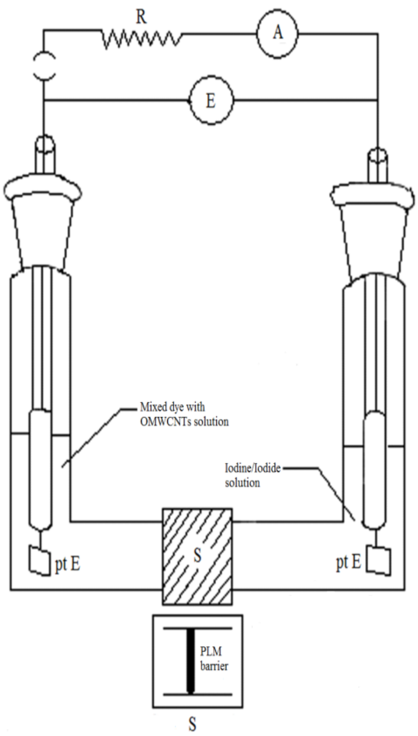

Figure 1. Schematic diagram of photoelectrochemical cell. S: PLM barrier, pt E - Electrode, E - electrometer, R - variable discrete resistance, A – ammeter.

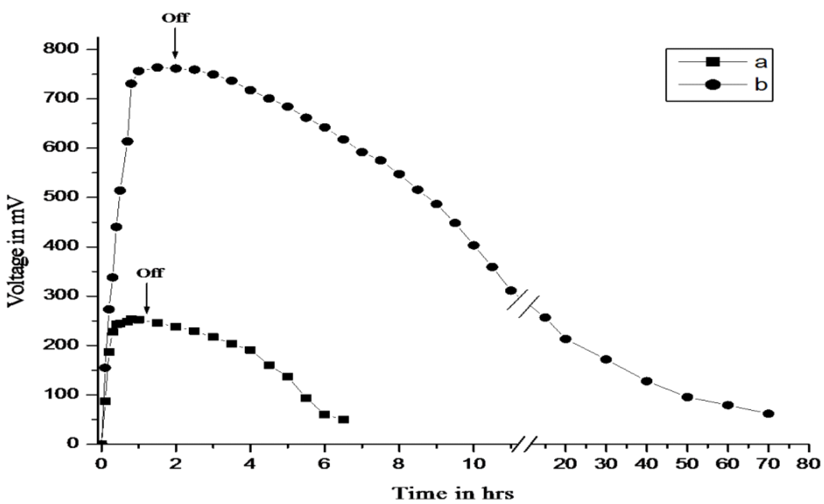

Fig 2. Growth and decay curve of photovoltage (V_{oc}) generation using planar lipid membrane as barrier for (a)PSF & AZC, (b)PSF & AZC adsorbed CNTs system.

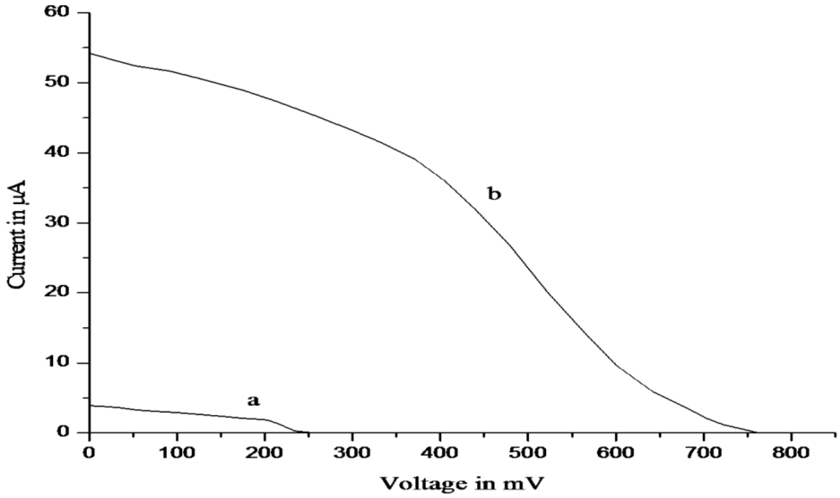

Fig 3. Current vs. voltage characteristic curves of the PEC cells for (a) PSF & AZC and (b) OMWCNTs adsorbed PSF & AZC.

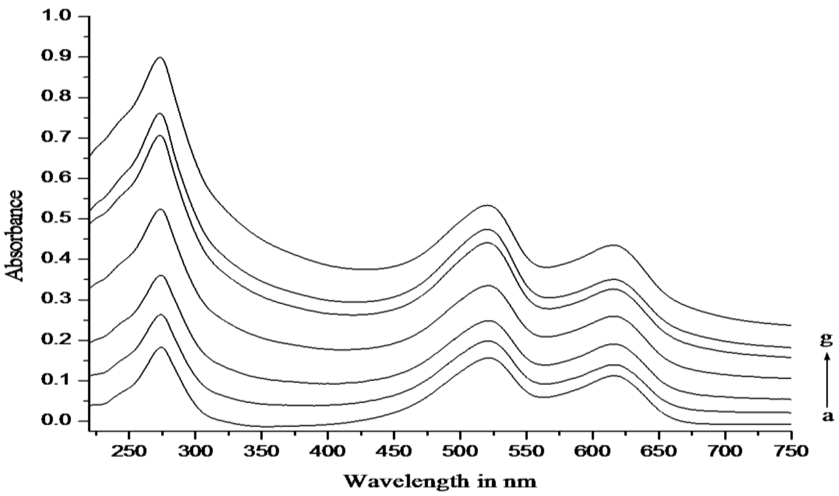

Fig 4. Absorption spectra of (a) PSF & AZC and (b to g) OMWCNTs adsorbed PSF & AZC systems. Concentration of PSF (a to g) 3 x 10^{-5}M, AZC (a to g) 3 x 10^{-5}M and OMWCNTs (x10^{-7} Kg/L) (a) 0.0, (b) 2.2, (c) 4.1, (c) 6.5, (d) 8.3, (e) 10.4, (f)11.1 and (g) 12.2.

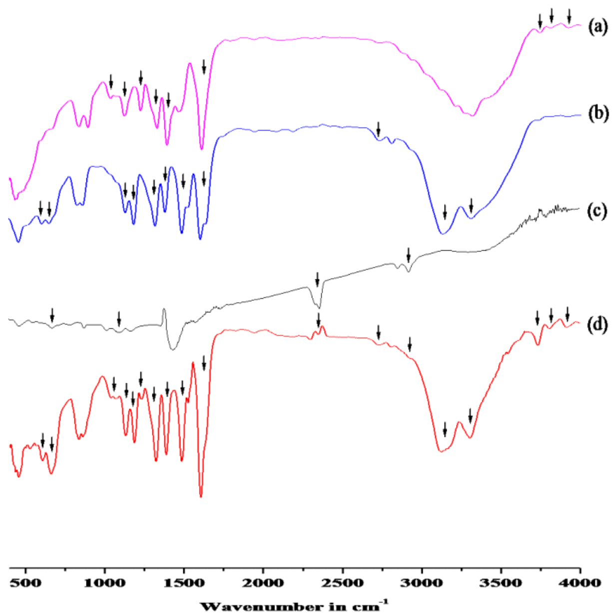

Fig 4. FITR spectra for (a) PSF, (b) AZC, (c) OMWCNTs, (d) Mixed system of PSF, AZC and OMWCNTs.

The PEC cell consisted of two L-shaped glass tubes (fig 1). A barrier was mounted on one tube which was fitted into the other by means of a standard joint. Here we have used a planar lipid membrane of oxidized cholesterol as barrier. One side of the barrier was bathed with Iodine Iodide (I^-/I^{3-}) solution and thve other side with aqueous solution in OMWCNT conjugated with PSF and AZC, in a 1:1 concentration ratio. A pair of platinum electrode was placed symmetrically across the barrier. A 60W lamp was used for illumination and the light intensity was measured by using a Luxmeter with photodetector (D & L Instrument, MS6610). Photovoltages and currents were measured by using Keithly digital multimeters (DM196). The energy conversion efficiency of the cell was calculated by using standard methods [8-10]. Keeping the cell under illumination, we recorded the photovoltage (Voc) generation.

Results and Discussion:

Upon illumination the photovoltage started increasing, attained a saturation value and remained constant till the illumination was there. When the light was switched off voltage started decreasing slowly (Fig 2). The storage time was quite high nearly 66 hrs. The photovoltage cycle was reproducible upon further illumination. From the current-voltage curve (Fig 3) we calculated the fill factor (FF) and energy conversion efficiency (η%) of the cells using equations (1) and (2).

FF = (V_{pp} x Ipp) / (V_{oc} x I_{sc}) ------------ (1)

η% = (Voc x Isc x FF x 100) / Incident light power -------------- (2)

The values of open circuit voltage (Voc), short circuit current (Isc), storage duration, fill factor (FF), energy conversion efficiency (η%) are summarized in the table . It is evident that the presence of OMWCNTs causes radical increase in the values of V_{oc}, I_{sc}, storage time, FF and η%.

. Spectral studies showed that with addition of OMWCNTs to a fixed concentration of PSF & AZC solution, the absorbance of PSF & AZC increased throughout the spectral range without any shift in the absorption peaks. The results clearly indicated that the dye molecules got adsorbed on OMWCNT side walls without any change in chemical as well as photochemical properties. Increase in the absorbance was a definite indication to the enhancement of the absorption of the incident photons which resulted in the amplification of efficiency of the cells [7]. One dimensional carbon nanotubes have excellent electron-storage capacity (one electron for every 32 carbon atoms) and exhibit metallic conductivity similar to metals [11].We propose that the presence of OMWCNT in the conjugate resulted in further improving the efficiency by improved charge transfer, charge transport and efficient charge collection [12].

Moreover, the hydrphobic barrier offered by the planar lipid barrier planar lipid membrane due to its low dielectric constant hindered back recombination of the photodissociated charges and as a result of which the efficiency as well as the storage duration of the cell increased further.

. We got maximum PV for OMWCNTs conc. (10.4 x10^{-7} Kg/L).

For further support to our proposed idea, we will perform FTIR of all sample. From Infrared Spectra, it can be suggested that the basic structure of the dyes remain changed as all the peaks from two dyes and OMWCNTs are found in the same position as previous, for the mixed system FTIR spectrum.

Conclusions

Our studies showed that the light-harvesting performance of such PEC cells was much better in presence of dye-CNT mixed systems. As a particular dye absorbed only a certain fraction of the incident light spectrum, being transparent to the rest, we used two different dyes to overcome the band absorption limits of each dye. The presence of OMWCNT boosted the absorption of incident photons. The use of lipid membrane barrier played an important role in enhancing the storage duration of the photovoltage and the overall efficiency of the cell. These low cost cells were easy to assemble compared to the intricate device fabrication techniques of other conventional solar cells

Conflicts of Interest

State any potential conflicts of interest here or “The authors declare no conflict of interest”.

References and Notes

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