Sensitization of Nanocrystalline Titanium dioxide Solar Cells using Natural Dyes: Influence of Acids Medium on Coating Formulation

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Abstract. Dye Sensitzed solar cells (DSSC) has drawn attention as an alternative to the silicon based solar cells due to their low fabrication cost and reasonably high efficiency. This paper presents an approach on the development of DSSC and the photoelectrochemical effect and conversion efficiency available natural dye sensitized Titanium dioxide (TiO₂) photoelectrode with respect to acid treatment of coating formulation. Acid treatment on TiO₂ paste was observed using an organic acid (citric acid) and an inorganic acid (nitric acid) considering a base natural dye (red spinach). Optimal concentration of these acids (organic and inorganic) was determined by cell performance. Five different natural dyes were used as sensitizers of DSSC and their performance was observed considering photoelectrochemical output and UV-visible absorption using optimized acid concentrations. Citric Acid treated cells with red spinach dye as sensitizer produced the best performance with 1mAcm⁻² current density with 505mV potential. It was found that DSSC showed better photovoltaic performance and higher conversion efficiency when the TiO₂ paste was treated by organic acid than inorganic acid and red spinach dye proved to be the best sensitizer.

Keywords: Dye Sensitized Solar Cell, Nanocrystalline, Natural Sensitizers, Photoelectrochemical output, Acid medium.

1. INTRODUCTION

Silicon based solar cells were the most popular before the emerging of dye-sensitized solar cells. These solid-state junction devices have dominated photovoltaic industry. Since Grätzel et al. developed dye-sensitized solar cells (DSSCs) in 1991, these have attracted a significant devotion due to their environmental friendliness and low cost of production along with reasonably high efficiency. A DSSC is composed of a nanocrystalline porous semiconductor (TiO₂) electrode-absorbed dye, a counter electrode, and an electrolyte containing iodide and tri-iodide ions. In DSSCs, the dye as a sensitizer plays a key role in absorbing sunlight and transforming solar energy into electric energy.

In conventional silicon based solar cells, silicon both absorbs the sunlight and acts as the charge carrier. But in DSSC, light is absorbed by the sensitizer and charge separation takes place by the photo-induced electron injection from dye to sensitizer. In contrast to conventional solar cells DSSC is a photoelectrochemical cell. It resembles natural photosynthesis in two respects like it uses organic dye based on chlorophyll to absorb lights and produces a flow of electrons. And it uses multiple layers to enhance both the light absorption and electron collection efficiency. The photoelectrode plays one of the most important roles in high efficiency of DSSC because this is the first step towards electron transport. So lots of efforts have been reported for the modification of the photo electrode.

Titanium dioxide (TiO₂) as a photocatalyst for degrading organic pollutions has attracted much attention because of its various advantages. Unfortunately, the technological use of TiO_2 is limited by its wide band gap (3.2eV for anatase), which requires UV light irradiation to obtain its photocatalytic activity. Because UV light only accounts for a small fraction (5%) of the sun's energy compared to visible light (45%), any attempt making TiO_2 absorb visible light will have a profound positive effect on its visible photocatalytic performance. For this purpose, various efforts have been directed toward the development of visible-light responsive TiO₂ materials. One was based on the chemical/physical adsorption of dye molecules on TiO₂ surface for construction of dye-sensitized photocatalysts that could work under visible light irradiation. Another approach utilized the chemical reaction between Ti-OH and phenolic hydroxyl of organic compounds (not dye), such as catechol, salicylic and binaphthol, to form surface complexes on TiO_2 surface and to realize visible light absorption. So-produced organic-modified TiO₂ was also a visible light activated photocatalyst. Previous studies showed that the concentration of acid has an impact on the performance of the cell. Thus far, several natural dyes have been utilized as sensitizers in DSSCs, such as cyanine, carotene, tannin, and chlorophyll.

Along with two acids (citric acid and nitric acid) five natural dyes from different sources was used in this work. These dyes were classified in two categories anthocyanin and chlorophyll based dyes. The chlorophyll based dyes were Red Spinach (Amaranthacea Gangeticus), Aurum Leaves (Colocasia Antiquorum) and Malabar Spinach Buds (Basella Alba). And the anthocyanin based dyes were Black Berry (Rubus Allegheniensis) and Black Grape (Vitis Vinifera). All of these dyes are available in large scale quite easily in Bangladesh. The aim of this project research work was to find out the effect of acid concentration on the photovoltaic performance of DSSC and also to find out an effective natural dye which gives the best performance as a sensitizer of DSSC.

2. EXPERIMENTAL

2.1 Materials

The materials used in this experiment were Indium Tin Oxide (ITO) coated glass plate (Dyesol, Australia), TiO₂ P25 Degussa (USA), Citric Acid ($C_6H_8O_7$), Nitric Acid (HNO₃), PEG, Titanium Isopropoxide (Merck, Germany), Triton X-100 (Merck, Germany), Ethanol (Germany) and Acetone (Germany).

2.2 Dye Extraction

The anthocyanin based natural dyes were extracted from Grapes and Black grapes using distilled water. They were crushed in distilled water and the filter solution was an anthocyanin solution.

The chlorophyll based natural dyes were extracted from Red spinach leaves, Malabar spinach buds and Aurum leaves by smashing them in a mortar and pastel using acetone. The solution was kept in a dark colour bottle for 2-3hrs. The filtered solution was a chlorophyll based dye.

2.3 TiO₂ electrode preparation

One gram of P25 Degussa was mixed with 1 ml acid (citric acid or nitric acid) and 0.5ml Titanium isopropoxide, then 0.5 ml Triton X-100 was added to the mixture and the total mixture was sonicated for 10 minutes so that uniform TiO_2 paste can be formed. Different concentrations of citric acid and nitric acid was used to make different types of TiO_2 paste keeping the above composition same to observe the cell performance in each case. Concentration used for citric acid were 1M, 0.75M, 0.50M, 0.3M, 0.2M, 0.1M, 0.075M and 0.05M the concentrations of nitric acid used were 0.5M, 0.4M, 0.3M, 0.2M, 0.1M, 0.08M, 0.08M, 0.06M, 0.04M, 0.02M, 0.01M, 0.003M, 0.001M, 0.003M& 0.0001M.

The prepared paste was coated on an ITO glass with a surface resistance of 10 ohms by using doctoral blading technique with an approximate thickness of 10-12 micrometre and the area of the cell was 1cm2. Then the coated plate was annealed at 400°C for 20 min. The TiO_2 coated glass plate was soaked in natural dye for 16hrs in a dark and sealed place. Then glass plate was washed using ethanol and dried in air for few minutes.

2.4 Electrolyte and counter electrode preparation

0.83g of 0.5M potassium iodide and 0.127g of 0.05M iodine was mixed in 10ml ethylene glycol. The solution was stored in a black bottle. This solution is used as the electrolyte.

The counter electrode was prepared by exposing the conductive side of an ITO coated glass to candle light for 2-3 minutes which leaves a dark shade of carbon on the glass

2.5 Cell fabrication and measurements

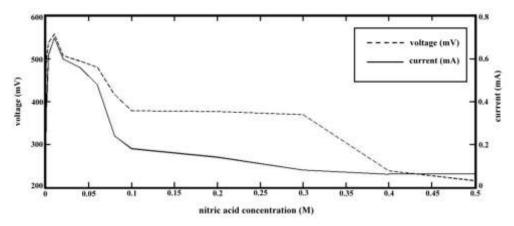
The two electrodes were combined together keeping TiO_2 paste coated surface and the carbon coated surface face to face. 15 micron of electrolyte solution was slowly given in the two glass contact and by the capillary action the electrolyte was uniformly distributed throughout the TiO_2 Nano crystals. When completed, the ready the DSSC was energized using a light source of a 55-W Xenon lamp mounted inside a solar simulator under illumination of 500W/m² (calibrated using a pyranometer). The lamp almost imitates spectrum of sunlight. The cell performance was measured using AGILENT 34401A precision multimeter. The testing temperature was 25°C-27°C. The UV visible absorbance was measured using T-60A spectrophotometer UV-Visible spectrophotometer (PG electronics U.K.).

3. RESULTS & DISCUSSION

3.1 Acid treatment outcome

The main idea of acid treatment of TiO_2 Nano particles was to see if it prevents Nano particles from coagulation and the Nano particles disperses into the paste solution well. In addition to, the good quality of the TiO_2 coating layer make lots of the chemisorption's site for the dyes. So cells were constructed with different acid concentration (HNO₃ and C₆H₈O₇). Red spinach was chosen as reference dye. Thus recorded the performance of the cells and then compared the results with a none acid treated cell. The results showed the enhanced energy conversion efficiency with the reduction of resistance at the interfaces between ITO substrate and TiO₂ Nano particles at optimum acid concentration. The result is shown in Fig.1, Fig.2 and Table-1.

Figure 1 and Figure 2 shows, acid containing paste formulated cells were enhanced of the current density in comparison with normal cell. In case of HNO₃ and $C_6H_8O_7$ treated cells showed higher short circuit current density, 0.7mAcm^{-2} and 1 mAcm^{-2} respectively, and reduced open circuit voltage of 559mV and 505mV respectively. None acid treated cells showed 0.5mAcm^{-2} short circuit current density and 0.75V open circuit voltage. These phenomena could be conferring that the acid contributed regular arrangement of the photo electrode by the dispersion of TiO₂ Nano particles. This dispersion is one of the factors to make much chemisorption site for the organic dyes. Also it proves that organic acid (citric



acid) treated cells yield more current and efficiency than inorganic acid (nitric acid).

Fig.1. Current density and voltage with the concentration of HNO₃

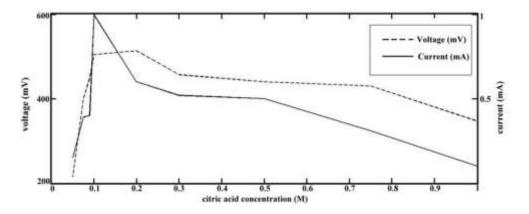


Fig.2: Current density and voltage with the concentration of $C_6H_8O_7$

Table 1 shows the supporting data that with proper acid treatment the conversion efficiency of the cells can be maximized. This actually indicates the fact that acid treatment improves the conductivity between ITO substrate and TiO_2 Nano particles by modifying powder surface. The photocurrent density and cell voltage properties of fabricated cell using 0.1M citric acid, 0.01M HNO₃ and none acid are shown in Fig.3 and proves that organic acid provides higher current gain than inorganic acid.

Table 1.	Performance	Summary

	Normal Device	HNO ₃	C ₆ H ₈ O ₇
$I_{sc}(mA/cm^2)$.5	.7	1
V _{oc} (V)	.75	.559	.505
FF(%)	39.4%	45.5%	57.76%
η(%)	0.2955%	0.3565%	0.583%

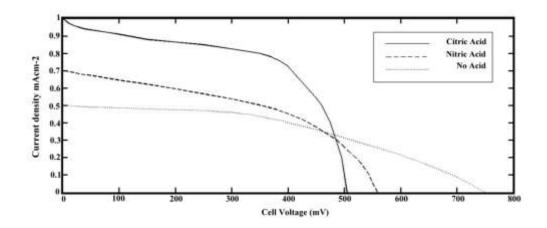


Fig.3: Photocurrent-photovoltaic (J–V) characteristics of acid treated TiO₂ nanoparticles of DSSCs Citric acid, Nitric acid and None Acid

3.2 Photoelectrochemical properties of DSSCs sensitized with natural dyes

Photovoltaic tests of DSSCs using these natural dyes as sensitizers were performed by measuring the current–voltage (I–V) under irradiation with white light (500Wm⁻²) from a solar simulator. The performance of natural dyes as sensitizers in DSSCs was evaluated by short circuit current density (J_{sc}), open circuit voltage (V_{oc}) and maximum absorbance wavelength (λ_{max}). The photoelectrochemical parameters of the DSSCs sensitized with natural dyes are listed in Table 2 and Table 3. Table 2 shows the natural dye performance considering 0.1 M citric acid treated TiO₂.

Dye Source	Solvent	λ_{max}	V _{oc} (mV)	J_{sc} (mAcm ⁻²)
Red Spinach	Acetone	440	505	1
Black berry	Distilled water	412	330	0.22
Black grape	Distilled water	427	269	0.1
Aurum Leave	Acetone	419	569	0.6
Malabar spinach bud	Acetone	483	386	0.31

Table 2: Citric acid (0.1M) for paste formulation	Table 2:	Citric acid	(0.1M) for	paste formulation
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Table 3 shows the natural dye performance considering nitric acid treated TiO₂. From these data it is evident that chlorophyll based dyes provide better electrochemical property than anthocyanin based dyes. Among the tested dyes, red spinach showed promising performance than any other dyes. Maximum short circuit current density of 1mAcm^{-2} was achieved using red spinach with citric acid treated TiO₂ electrode. Next mentionable dye performance was seen by aurum leave extracted dye. It provided 0.6mAcm⁻² short circuit current for citric acid

treated TiO_2 electrodes. Even in these tests we found that organic acid (citric acid) yields better result than inorganic acid (nitric acid).

Dye Source	Solvent	λ_{max}	V _{oc} (mV)	J _{sc} (mAcm ⁻²)
Red Spinach	Acetone	440	559	0.7
Black Berry	Distilled water	412	248	0.11
Black grape	Distilled water	427	296	0.02
Aurum Leave	Acetone	419	580	0.31
Malabar spinach bud	Acetone	483	370	0.22

Table 3:HNO₃ (0.01M) for paste formulation

3.3 UV-visible absorption spectroscopy of natural dyes

As mentioned earlier five different natural dyes from five different sources were selected as sensitizers. To understand their potency, UV-vis light absorption was observed. So their extraction used acetone and distilled water respectively. Since no other purification method was used extracted dyes were in crude form. Then their UV-vis light absorption was measured. Figure 4 and Figure 5 shows the absorbance with respect to wavelength of light for chlorophyll based and anthocyanin based dyes respectively. From Table 2 and 3 and Fig. 4 and 5 some explanation can be made. Since red spinach has a broader spectrum (312nm-698nm) and higher absorption, it provided better conversion efficiency than other dyes. The reason for aurum leave dye to give second best result is because of its absorbance over a broad spectrum (309nm-750nm) but comparatively lower amount of absorbance. Malabar spinach gave absorbance in the range of 396nm-616nm bandwidth which is narrower compared to red spinach so it has lesser photelectrochamical output. For anthocyanin based dyes, black berry (320nm-540nm) dye seemed to give better photoelectrochamical output for its wider bandwidth than black grape dye (342nm-496nm).

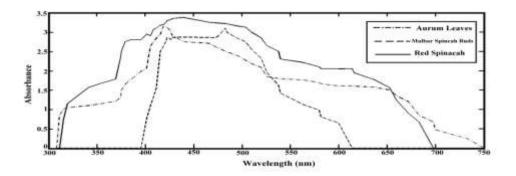


Fig.4: Chlorophyll Based Dye UV-vis Response

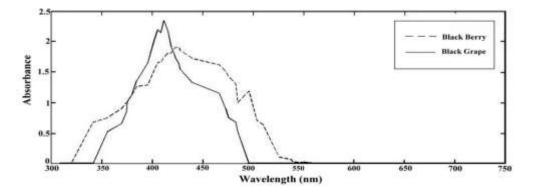


Fig.5: Anthocyanin Based Dye UV-vis response

3. CONCLUSION

Maximum efficiency conditions related with the adhesive properties and conversion efficiency were achieved by the variation of the acid and concentrations. In comparison with acid untreated DSSC, the enhancement of conversion efficiency and fill factor were significant enough to consider. The better acid effect was judged and the optimal amount was found. Acid treated DSSCs showed increased current density and fill factor, but open circuit voltage was slightly reduced. These studies provide proof for the dependency of acid treatment on better TiO_2 electrode for DSSC purpose. On the other hand the photoelectrochamical performance of 5 natural dyes revealed the potential of red spinach as sensitizer and which is locally available in Bangladesh. Overall, natural dyes as sensitizers of DSSCs are promising because of their environmental friendliness, low-cost production.

Acknowledgements:

This research work is funded by Ministry of Science & Technology, Bangladesh Government. The authors thank Dr. Firoz Alam Khan of BUET material science lab for his help & support on numerous occasions.

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Mubarak A. Khan Experience in fiber reinforced polymer composite materials for various applications such as parts & body of auto car, panelized constriction materials, bodies of electric appliances. Totally biodegradable composite materials based on natural fibers and degradable (both and synthetic) thermoplastic and resin for biomedical purposes. Also experience in radiation processing technology for biomedical purposes, modification of natural fibers, stimuli-responsive materials form natural polymers. Worked in Germany (Technical University of Berlin, Fraunhofer Institute of Applied Polymer Research) as DAAD and AvH fellow, in Japan as MIF Fellow, in USA (Michigan State University) as visiting scientist, in Australia (University of New South Wells) as IAEA fellow. Part time and visiting Professor and Examiner of various universities of Bangladesh. Served as project director/co-project director of different national and international scientific project on polymer science. Reviewers of different International Journals on polymer and composite and natural fiber modification. Jute based polymer composites are developed and are being using commercially in Bangladesh for housing materials (Jutin).

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Sabia Sultana biography not available

Jahid M M Islam biography not available

Jasim Uddin biography not available