

## Preparation and Characterization of Dye Sensitized Solar Cell Using Natural Dye Extract from Red Amaranth (*Amaranthus sp.*) as Sensitizer

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Received: 26 Feb. 2015, Revised: 7 Apr.2015, Accepted: 8 Apr.2015.

Published online: 1 May 2015.

**Abstract:** Dye Sensitized Solar Cells (DSSCs) were assembled with crystalline titanium dioxide (TiO<sub>2</sub>) coating on a conductive glass (Indium doped tin oxide, ITO) and absorbed with a novel natural dye extracted from red amaranth (*Amaranthus sp.*). Solvents of various polarities such as water, ethanol, methanol and acetone were used to extract the dye. Dye sensitization time of TiO<sub>2</sub> electrode and solvent for extracting dyes were optimized considering photoelectric output. Again, different polar portions of crude dyes were separated using various polar solvents by means of thin layer and column chromatography technique. The best light to electricity conversion efficiency was obtained when sensitization time of electrode was 30 min and dyes were extracted by acetone in crude form. Sample showed maximum cell voltage 0.492 V, current density 0.78 mA/cm<sup>2</sup> and cell efficiency 0.22%.

**Keywords:** Dye Sensitized Solar Cell; Sensitizer; Natural dye; Cell efficiency; Dye sensitization time.

### 1 Introduction

World's conventional energy supplies (oil, natural gas, and coal) have a limited lifetime. So alternative source of energy is needed and dye-sensitized solar cells (DSSCs) are promising alternative for future large scale power production from renewable energy sources. As a novel renewable and clean solar-to-electricity conversion system DSSCs are considered viable substitutes to the conventional silicon-based photovoltaic cells since the pioneering work reported by O'Regan and Grätzel [1]. DSSCs are expected to be used for future clean energy due to their low fabrication cost, simple preparation technique and environmental friendliness. DSSCs are made of low-cost materials and do not need elaborate apparatus to manufacture. Generally DSSCs contain mainly five components: (1) a conductive and transparent mechanical support, (2) a wide band gap semiconductor film, (3) a sensitizer, (4) redox couple (usually I<sub>3</sub><sup>-</sup>/I<sup>-</sup>) in an organic solvent, and (5) a counter electrode.

The conversion of the absorbed photon to electrical current in DSSCs is achieved by injecting electrons from the photoexcited dyes into the conduction band of the photoelectrode. Thus, the sensitizing dye plays an important role in the absorption of the photons

and generation of electron and hole pairs; the charge carriers are transferred to the photoelectrode and the electrolyte. It is well known that the number of sensitizing dye molecules adsorbed on a photoelectrode is one of the key factors in determining the short circuit current density (J<sub>sc</sub>) and hence the efficiency in DSSCs [2]. Among the dye-sensitized solar cells, the DSSCs using ruthenium complex show a conversion efficiency of about 11%–12% [3-5]. However, ruthenium is a rare metal, so the cost of the ruthenium complex is very high. Use of these expensive Ru metals, derived from relatively scarce natural resources, corresponds to a relatively heavy environmental burden [6]. On the other hand, natural dye is easy to obtain, and its cost is very low and it can be used for same purpose [7]. In order to replace the expensive ruthenium compounds, many kinds of organic synthetic dyes have been actively studied and tested as low-cost materials [8, 9]. In nature, some fruits, flowers, leaves and so on show various colours and contain several pigments that can be easily extracted and then employed in DSSC [10]. Hence, it is possible to use natural dyes as alternative photosensitizers with appreciable efficiencies and their advantages over synthetic dyes include easy availability, abundance in supply, can be applied without further purification, environmentally friendly and they considerably reduce the cost of devices [11]. Zhou et al. [12] reported on twenty dyes obtained from nature, including flowers, leaves of plants, fruits, traditional

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Chinese medicines, and beverages were used as sensitizers in DSSC. The dyes extracted from these materials contained cyanine, carotene, chlorophyll, etc. The photoelectrochemical performance of the DSSCs based on these dyes showed that the open circuit voltages ( $V_{oc}$ ) ranged from 0.337–0.689 V, and the short circuit photocurrent densities ( $J_{sc}$ ) ranged from 0.14–2.69 mA  $cm^{-2}$ . In the majority of previous studies, the electrodes were immersed in dye for 12 h – 24 h to ensure titanium dioxide ( $TiO_2$ ) particles were completely covered with dye molecules [12-14]. Chien et al. [15] investigated the effects of the time of  $TiO_2$  film immersed in the anthocyanin extract on the performance of DSSC and found that  $V_{oc}$  and fill factor (FF) of longer immersion time were slightly higher. On the other hand,  $J_{sc}$  and cell efficiency ( $\eta$ ) steadily increased with immersion time and reached the maximum at 15 min. It was reported that the extracting solvent has an effect on the efficiency of DSSC [16, 17]. Chien et al. [15] also investigated the effects of purified and unpurified anthocyanin as sensitizer of DSSC.

In our previous study Khan et al. [14] investigated five natural dyes (red amaranth, black berry, black grape, arum leaf and malabar spinach bud) and found red amaranth has a good photoelectric performance as a sensitizer. Red amaranth (*Amaranthus sp.*) is commonly known as lalshak and it is available in Bangladesh all the seasons. The main aim of this work is to optimize various conditions for fabrication of DSSC using red amaranth dyes and to find out exact solvent system for extracting red amaranth for better output. In this paper dye sensitization time of red amaranth has been optimized. Using solvents of various polarities (normal water, boiling water, ethanol, methanol, and acetone) suitable solvent for extracting dyes have been chosen. Crude and different portion of dyes were used finding maximum cell efficiency. SEM images have been taken to observe the surface morphology. Extracted dyes have been characterized by UV-Vis absorption spectra.

## 2 Experimental

### 2.1 Materials

Materials used in this experiment were Indium Tin Oxide (ITO) coated glass plate (Dyesol, Australia),  $TiO_2$  (Degussa P25, USA), Citric acid ( $C_6H_8O_7$ ), Polyethylene Glycol (PEG), Titanium IV Isopropoxide (TTIP) (Merck, Germany), Triton X-100 (Merck, Germany), Acetone (BDH, UK), Ethanol (BDH, UK), Methanol (BDH, UK), Dye extracted from red amaranth (*Amaranthus sp.*, local Bangladeshi name lalshak) in our lab, Carbon (tip of candle flame), Potassium, Iodide, and Iodine.

### 2.2 Extraction of natural dye

Red amaranth (*Amaranthus sp.*) leaves were collected from local market and washed with water and kept them

some times in room temperature to remove the surface water. 100g of leaves were weighted and crushed adding 50 mL solvent. The extracted dyes were filtered three times with cotton cloth and stored in dark bottle covered with aluminium foil paper. To observe the effect of solvent five different solvents of various polarities such as normal water, boiling water, ethanol, methanol, and acetone were used for extraction dye from red amaranth.

### 2.3 Separation of dye

Crude dye from red amaranth was partially purified into different pigments using thin layer chromatography (TLC) and column chromatography. Three different colored pigments have been found in the TLC paper.  $R_f$  values for the pigments are 0.5952, 0.2381, and 0.00 for yellow, green, and red color respectively. TLC determined the dichloromethane and acetone mixture with ratio 4:1 as mobile phase. Later on column chromatography was done to separate these three different colored pigments. As red colored pigment showed negligible value so polar solvent methanol: acetone with ratio 4:1 was used only to collect it. All the pigments then concentrated at 25<sup>o</sup> C by rotatory vacuum evaporator and then collected.

### 2.4 Preparation of $TiO_2$ electrode

2 g of Degussa P25 powder was deposited in a beaker mixed with 6 mL citric acid of 0.1 M, 0.2 mL polyethylene glycol, 0.2 mL titanium (IV) isopropoxide and 0.1 mL nonionic surfactant triton X-100. The mixture was well mixed with the help of a glass rod and then kept in an ultrasonic bath for half an hour for the production of suspension with a consistency of a thick paint produced. The prepared paste was coated on ITO glass with surface resistance of 10-30 ohms by using doctor blade technique with an approximate thickness of 40  $\mu m$  [18] and the area of the cell was 1  $cm^2$ . Then the electrodes were sintered at temperature 450<sup>o</sup> C for half an hour [18]. After the annealing was completed, the  $TiO_2$  coated conductive glass was allowed to cool slowly at room temperature. The  $TiO_2$  coated glass plate was soaked in red amaranth dye and the soaking time was varied (10 min, 20 min, 30 min, 1 h, 2 h and 18 hours) and was kept at a dark place. The glass plates were washed with water and then ethanol and were kept in air for 30 minutes in room temperature to remove surface water.

### 2.5 Preparation of Electrolyte and counter electrode

8.3 g of 0.5 M potassium iodide and 1.27 g of 0.05 M iodine was mixed in ethylene glycol until it is 100 mL. The solution was stored in a black bottle and was used as electrolyte. For the preparation of counter electrode a light carbon film was applied for 20 sec to the conductive side of the ITO glass from the tip of candle flame.

## 2.6 Assembling the Cell

Electrode and counter electrode were combined together keeping TiO<sub>2</sub> paste coated surface and the carbon coated surface face to face. 2/3 drops of electrolyte solution was given in the contact of two glasses and by the capillary action the electrolyte was uniformly distributed throughout the stained TiO<sub>2</sub> film. Excess electrolyte from the exposed area of the glass was wiped off by using cotton or tissue. The complete cell was then taken to sunlight for illumination.

## 2.7 Characterization and measurement

Surface morphology and crystalline structure of the TiO<sub>2</sub> film was analysed by SEM (model XL 30, Philips, ESMX). The absorption spectra of dye solutions were recorded using UV-Vis spectrophotometer (model T-60A, PG electronics U. K). Electrical properties were measured by using two digital multimeter keeping the cell in sunlight of approximately 100 mW/cm<sup>2</sup> illumination. Current and voltage were measured by multimeters changing resistance with the help of a variable resistor. Based on current density-voltage (J-V) curve, the fill factor (FF) is defined as

$$FF = (J_{\max} \times V_{\max}) / (J_{sc} \times V_{oc}) \quad [19]$$

Where  $J_{sc}$  and  $V_{oc}$  are the short-circuit current density (mA/cm<sup>2</sup>) and open-circuit voltage (V) respectively,  $J_{\max}$  and  $V_{\max}$  are the short-circuit current density and open-circuit voltage at maximum power point ( $P_{\max}$ ) of the J-V curve. The overall energy conversion efficiency ( $\eta$ ) is defined as

$$\eta = (J_{sc} \times V_{oc} \times FF) / P_{in} \times 100\% \quad [19]$$

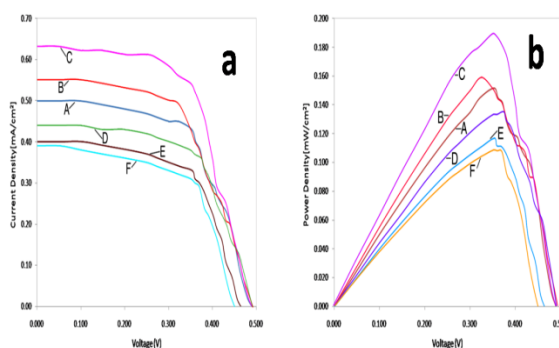
Where  $P_{in}$  is the power of incident light.

## 3 Results and Discussion

### 3.1 Effect of dye sensitization time of electrode

Dye sensitization process time is the important parameter to increase the overall performance of the DSSC. In this experiment acetone has been used for the extraction of red amaranth dye with the reference of Khan et al. [14] and the sensitization time of electrode into dye has been varied. The dye sensitization times have been varied for 10 min; 20 min; 30 min; 1 h, 2 h and 18 hours. The TiO<sub>2</sub> nanocomposite shows maximum performance at the sensitized time of 30 min and then decreases accordingly with the increasing time. **Fig 1** shows the current density-voltage (J-V) and power-voltage (P-V) characteristic curves of the prepared DSSC that takes red amaranth

extract as the natural dye for immersion time of 10 min to 18 hours. **Table 1** shows the efficiency of DSSC is 0.15% for the immersion time of 10 min, 0.16% for 20 min, 0.19% for 30 min, 0.14% for 1 hour, 0.12% for 2 hour and 0.11 % for 18 hour. As shown in table 1 with sensitization time of 30 min the conversion efficiency ( $\eta$ ) of the DSSC is 0.19%, with open-circuit voltage ( $V_{oc}$ ) = 0.490 V, short-circuit current density ( $J_{sc}$ ) = 0.63 mA/cm<sup>2</sup>, maximum power ( $P_{\max}$ ) = 0.189 mW, and fill factor (FF) = 0.61.  $J_{sc}$ ,  $P_{\max}$  and  $\eta$  three major parameters of DSSC have been increased with time and for the sensitization time of 30 min these parameters were maximum. For the longer immersion time  $J_{sc}$ ,  $P_{\max}$  and  $\eta$  have been decreased. Here the decrease in the efficiency ( $\eta$ ) after 30 min may be due to dissolved the dye into solvent again which has been absorbed into the surface of the TiO<sub>2</sub> nanostructures. The result shows that the absorption of red amaranth dye by Degussa P25 is faster than many other dyes which required 12h-24h [12-14].



**Fig.1.** (a) Current density-Voltage (J-V) and (b) Power-Voltage (P-V) curves for the DSSC with sensitization time of electrode into dye for (A) 10 min, (B) 20min, (C) 30 min, (D) 1 hour, (E) 2 hour, and (F) 18 hour.

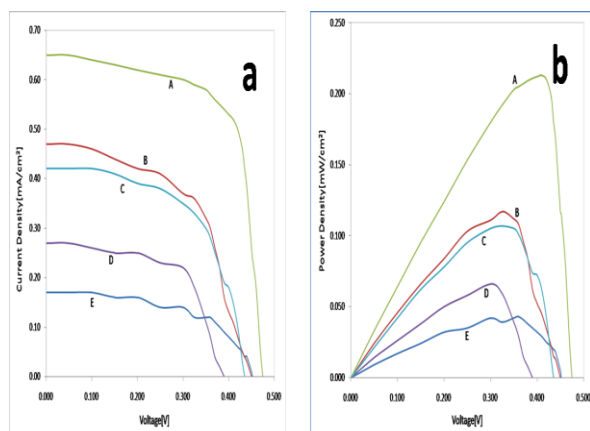
**Table 1:** Photoelectric parameters of DSSC for different sensitization time of electrode into dye.

Time	$V_{oc}$ [V]	$J_{sc}$ (mA/cm <sup>2</sup> )	$P_{\max}$ [mW]	F.F.	$\eta$ %
10 min	0.490	0.50	0.152	0.62	0.15
20 min	0.492	0.55	0.159	0.59	0.16
30 min	0.490	0.63	0.189	0.61	0.19
1 hour	0.491	0.44	0.135	0.62	0.14
2 hour	0.465	0.40	0.117	0.63	0.12
18 hour	0.450	0.39	0.109	0.62	0.11

### 3.2 Effect of solvent for extracting dye from red amaranth.

In this experiment different solvents of various polarities have been used to investigate the effect on the performance of DSSC keeping the optimized dye sensitization time 30 min constant. **Fig 2** shows the J-V and P-V characteristic curves of the prepared DSSC with red amaranth dye extracted by different solvents. As shown in **table 2** the conversion efficiency of DSSC is

0.04% for the solvent of water at room temperature, 0.12% for boiling water, 0.07% for ethanol, 0.11% for methanol and 0.21% for acetone. Table 2 shows that when the solvent is acetone to extract dye from red amaranth the conversion efficiency ( $\eta$ ) of the DSSC is maximum, with open-circuit voltage ( $V_{oc}$ ) = 0.475V, short-circuit current density ( $J_{sc}$ ) = 0.65 mA/cm<sup>2</sup>, and fill factor (FF) = 0.69. Table 2 shows that dyes extracted by acetone obtained all parameters of DSSC greater than other four solvents. So this experiment proves the dependency of solvent effect and its reveals solvent of less polarity such as acetone, is more capable to extract active portion of red amaranth dye than other four solvents.



**Fig.2.** (a) Current density-Voltage (J-V) and (b) Power-Voltage (P-V) curves for the DSSC extracted by (A) acetone, (B) boiling water, (C) methanol, (D) ethanol, and (E) water of room temperature.

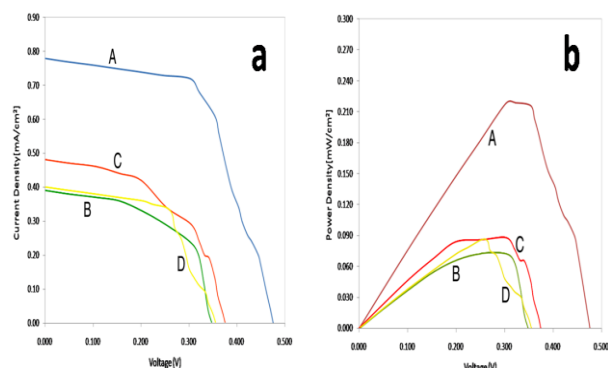
**Table 2:** Photoelectric parameters of DSSC for different solvents to extract dye from red amaranth.

Solvent	$V_{oc}$ [V]	$J_{sc}$ (mA/cm <sup>2</sup> )	$P_{max}$ [mW]	F.F.	$\eta$ %
water of room temp	0.452	0.17	0.043	0.56	0.04
Boiling water	0.450	0.47	0.117	0.55	0.12
Ethanol	0.390	0.27	0.066	0.63	0.07
Methanol	0.435	0.42	0.107	0.59	0.11
Acetone	0.475	0.65	0.213	0.69	0.21

### 3.3 Effect of Crude and Separated dye

In this experiment crude and different portions of separated dyes extracted from red amaranth were taken to compare the photoelectric performance. Here solvent was acetone for the extraction of dye and sensitization time was 30 min. Crude dye was extracted and then it was filtered after then it was separated using TLC and column chromatography and three distinct different coloured (green, yellow, and red) dyes were collected and used as sensitizer. **Fig 3** shows the J-V and P-V characteristic curves of the prepared DSSC sensitized with crude and separated red amaranth extract as the natural dye. **Table 3** shows that efficiency of DSSC is 0.22% for crude dye,

0.07% for green coloured dye, 0.09% for both red and yellow coloured dye. As shown in table 3 the conversion efficiency ( $\eta$ ) of the DSSC is maximum for crude dye, with open-circuit voltage ( $V_{oc}$ ) = 0.476 V, short-circuit current density ( $J_{sc}$ ) = 0.78 mA/cm<sup>2</sup>, and fill factor (FF) = 0.59. Table 3 shows that in case of crude dye every parameters of DSSC are greater than three other separated dyes. So explanation may be given that there are some unknown components in crude extract that could help dye molecules in the conversion of light to electricity. Chien et al. [15] used purified and crude anthocyanin extract as sensitizer and found that the performance of the cell with purified dye was worse than that with crude dye in all parameters.

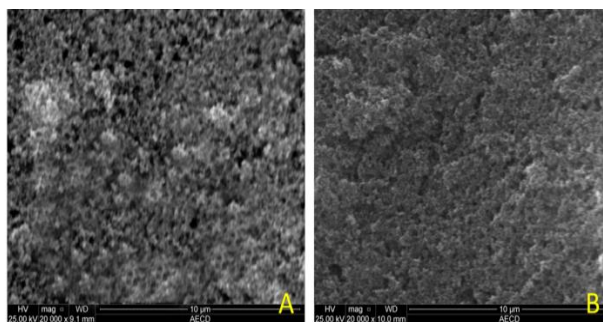


**Fig.3.** (a) Current density-Voltage (J-V) and (b) Power-Voltage (P-V) curves for the DSSC with red amaranth extract of (A) crude dye (B) green coloured dye (C) red coloured dye (D) yellow coloured dye.

**Table 3:** Photoelectric parameters of DSSC for crude dye and separated dye extract from red amaranth.

Dye	$V_{oc}$ [V]	$J_{sc}$ (mA/cm <sup>2</sup> )	$P_{max}$ [mW]	F.F.	$\eta$ %
Crude Dye	0.476	0.78	0.219	0.59	0.22
Green colour	0.348	0.39	0.073	0.54	0.07
Red colour	0.375	0.48	0.088	0.49	0.09
yellow colour	0.355	0.40	0.086	0.61	0.09

### 3.4 Morphological analysis of the surface



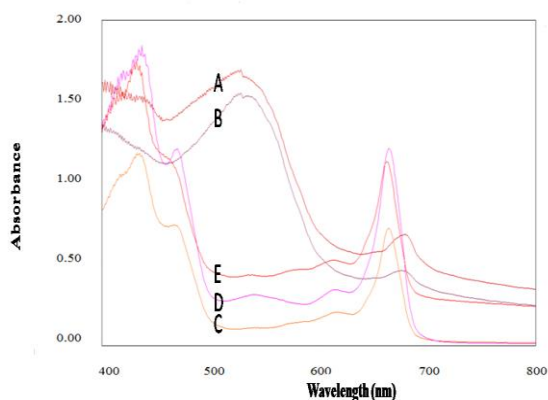
**Fig.4.** SEM images of Degussa P25 (A) before, and (B) after sensitization into dye.



**Fig. 4** showed the scanning electron microscope (SEM) image of the Degussa P25 coating on ITO annealed at 450° C. It was found that sample showed uniform crystalline nanostructure with evenly distributed pores ranged 900-1000 nm. No distinguishable change was found between the samples with dye and without dye. Both the samples showed rough surfaces which may be formed due to uneven manual coating. These rough surfaces probably responsible for increased electrical resistance resulting reduced efficiency. It can be assumed that same formulation will produce cells with higher efficiency if coated by a precise automatic coating machine.

### 3.5 UV-Vis spectroscopy analysis

**Fig. 5** shows the representative UV-Vis absorption spectra of dyes extracted from red amaranth using different solvents of various polarities. It is found that dyes extracted by water of room temperature and boiling water both have absorption peak at 680 nm (Fig. 5A, B) corresponds to the energy of 1.83 eV and another higher absorption peak is 520 nm corresponds to the energy of 2.39 eV. Again, dyes extracted by ethanol, methanol, and acetone have absorption peak at around 665 nm (Fig. 5C, D, E) corresponds to the energy of 1.87 eV and another higher absorption peak at around 430 nm correspond to the energy of 2.89 eV.



**Fig.5.** UV-Vis absorption spectra of red amaranth dyes extracted by different solvents of various polarities such as (A) water of room temperature, (B) boiling water, (C) ethanol, (D) methanol, and (E) acetone.

The energy associated with the longer wavelengths corresponds to lower energies are not sufficient to overcome the band-gap of P25 (3.1eV) which has been used on photo anode as semiconductor oxide. So, it can be assumed that extracted dyes having absorption peak of longer wavelengths will not be a good sensitizer. On the other hand, the energy associated with the absorption peak (430 nm) of ethanol, methanol, and acetone extracted dyes were near similar to the bandgap energy of Degussa P25. So, mostly the energy associated with second absorption peaks are responsible for the

transition of electron from the highest occupied molecular orbital (HOMO) to the lowest unoccupied molecular orbital (LUMO). Though ethanol, methanol, and acetone extracts showed almost same absorption peaks but from table 2 it was found that acetone extract showed maximum cell efficiency which suggested that the pigment which responsible for the peak around 430 nm in acetone extract have higher electron splitting ability compared to the pigments of ethanol and methanol extract.

## 4 Conclusion

The main objective of this study was to make a cost effective dye sensitized solar cell (DSSC). For this purpose this study explored three key parameters towards the optimization of DSSC. The conclusion reports that an efficient natural dye from red amaranth (*Amaranthus sp.*) which is locally available in Bangladesh has been identified and its maximum cell efficiency was 0.22%. This study reported that (a) optimized dye sensitization time for red amaranth was 30 min, (b) acetone performed as a good solvent for dye extraction, and (c) crude extract was better than further separated dye. Although the efficiency obtained with red amaranth dyes are below the requirements for large scale production, the results are very promising and believed to produce higher performance if cells were coated more uniformly and precisely. With the advantages such as ready availability, quick and easy preparation procedures and environmental friendliness DSSC solar cell sensitized with this natural dye deserves further exploration.

## Acknowledgments

This work was funded by Ministry of Science and Technology, Government of Bangladesh and supported by the University Grand Commission of Bangladesh and Ministry of Education, Bangladesh. I would like to acknowledge Mahmudul Hasan Khan (M.Sc. Student, Department of Chemistry, Jahangirnagar University Savar, Dhaka, Bangladesh) for TLC and column chromatography.

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