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UTILIZATION OF NATURAL HENNA IN COMBINATION WITH BLACK TEA AS A SENSITIZER IN DYE SENSITIZED SOLAR CELLS

C. N. Nupearachchi* and V. P .S. Perera

Department of Physics, The Open University of Sri Lanka, Nawala, Nugegoda

INTRODUCTION

Solar cells have drawn interest as efficient devices to produce electricity from sunlight. With the development of material science and engineering, various devices have been fabricated to replace the expensive silicon based solar cells. Among them, one of the promising device is the low cost and high efficient dye sensitized solar cell (DSSC) with simple assemble technology which was developed by Micheal Grätzel at EPFL in 1990s (O'Regan and Gratzel, 1991).

DSSCs are a combination of materials, consisting of a transparent mesoporous film of nanocrystalline semiconductor electrode coated with a dye, an electrolyte containing a suitable redox couple and a Pt sputtered counter electrode (Longo and De Paoli, 2003).

The use of sensitizers which has a broad absorption band in conjunction with oxide semiconductor films of nanocrystalline morphology permits to harvest a fraction of sunlight. The illumination leads the dye to its excited state which is quenched by the electron transfer to the conduction band (CB) of the semiconductor, leaving the dye in an oxidized state. The oxidized dye is reduced by the electron donor, which is present in the electrolyte. The collected electrons in the CB flow through the external circuit to the counter electrode where they cause the reverse reaction of the redox mediator. Attention has drawn for the use of natural pigments in DSCCs as the dye sensitization is a process similar to the natural photosynthesis. Several natural pigments from flowers, fruits, leaves etc. have been used as sensitizers of DSSCs because of their ability to inject electrons from excited pigments to the semiconductor electrode.

The leaves of henna plant (*Lawsonia inermis*) have a red-orange dye, napthaquinone. Normally, it has been used as a hair dye over centuries. In this study, a DSSC was fabricated using the extract of henna leaves in combination with black tea as the sensitizer. The efficiency of this solar cell is compared with a cell made from henna alone as the sensitizer of DSSCs.

METHODOLOGY

TiO_2 films (1 cm × 1 cm) of thickness 10 μm were prepared using doctor blade method on conducting tin oxide (CTO) glass plates (15 Ωcm^{-2}) which was made grinding Degussa P25 powder with acetic acid and ethanol. Dried films were sintered at 450 ^{0}C in a furnace for 30 minutes.

First, 1.5 g of dried leaves powder of henna was added to 50 ml of water. After that, filtrate of 3.0 g of black tea boiled in 50 ml of water was mixed with this henna solution. The mixture was stirred maintaining constant temperature of 100 ^{0}C for 30 minutes. Simultaneously, a separate henna solution was made by adding 1.5 g in 50 ml of water at 100 ^{0}C. Then both the solutions containing henna alone and (henna + black tea) were filtered. TiO_2 films were immersed separately in the filtrates for 15 hours. Dye sensitized solar cells were fabricated by attaching a Pt sputtered CTO glass plate on the top of the dye coated electrode as the counter electrode. The capillary space in between was filled with an electrolyte containing $KI + I_2$ ($KI = 0.5$ mol dm^{-3}, $I_2 = 0.05$ mol dm^{-3}) which was dissolved in ethylene carbonate and acetonitrile in 4:1 ratio. Cells

* All correspondence should be addressed to Miss. C. N. Nupearachchi.(e-mail: chathunilnupe@gmail.com)

were characterized by illuminating with tungsten filament light (100 mW cm^{-2}) and a galvanostat / potentiostat coupled to a computer. Illumination was carried out through the TiO_2 layer of the cell. Absorption spectra were obtained with the Shimadzu UV-3000 spectrometer.

Also, the complex plane impedance spectra of these films were analyzed by Solartron 1260 frequency response analyser using SMART software provided with the instrument. A sweep was carried out for these films coated on CTO glass with Pt as the counter electrode by setting AC level at 500 mV in the frequency range from 1 MHz to 1 Hz while measuring the impedance in 1.0 s integration.

RESULTS AND DISCUSSION

The pigment extracted from henna leaves mainly contains hennatannic acid or 2-hydroxy-1, 4-napthaquinone which is red-orange in colour. The hydroxyl group of the hennatannic acid reacts with the Ti^{4+} ions on the surface of the TiO_2 film. Thus, the pigment can readily chelate with the surface Ti^{4+} ions by eliminating a water molecule as shown in Figure 1.

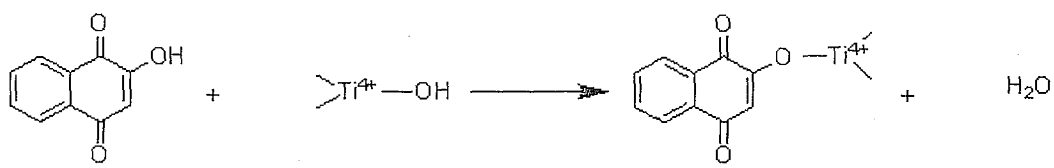

Figure 1. Mechanism of chelation of hennatannic acid with TiO_2

Black tea has many amino acids and more than 60% of the amino acid content is Theanine. It reacts with henna according to Michael addition as shown in Figure 2.

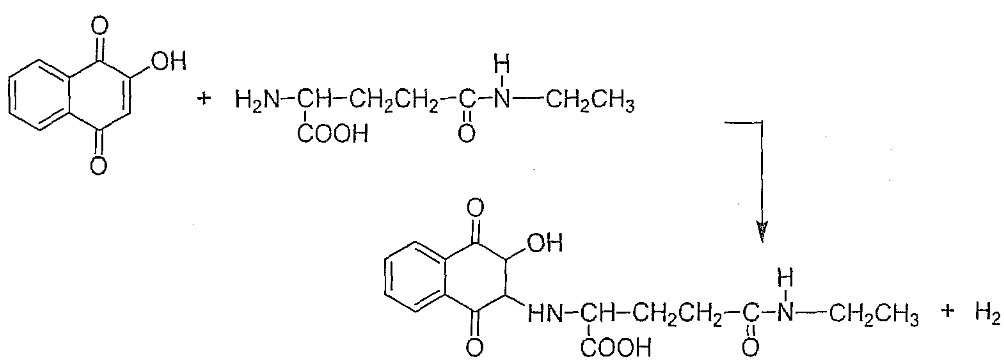

Figure 2. Michael addition of Theanine and henna

The resultant structure will have a COOH group which can react with the surface Ti^{4+} ions by eliminating a proton as in Figure 3.

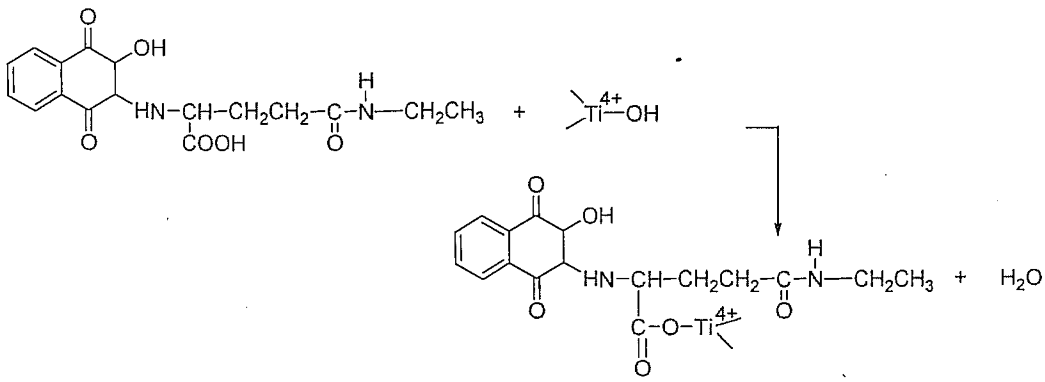

Figure 3. Mechanism of chelating with TiO_2 by (henna+ Theanine)

In the conjugated chromophores, the electrons jump between energy levels that are extended π orbitals, created by a series of alternating single and double bonds in aromatic systems.

Absorption spectrum of the pigment extracted from henna leaves and black tea in aqueous solution is shown in Figure 4. It has exhibited an intense absorption band in the UV region with peaks around 300 nm.

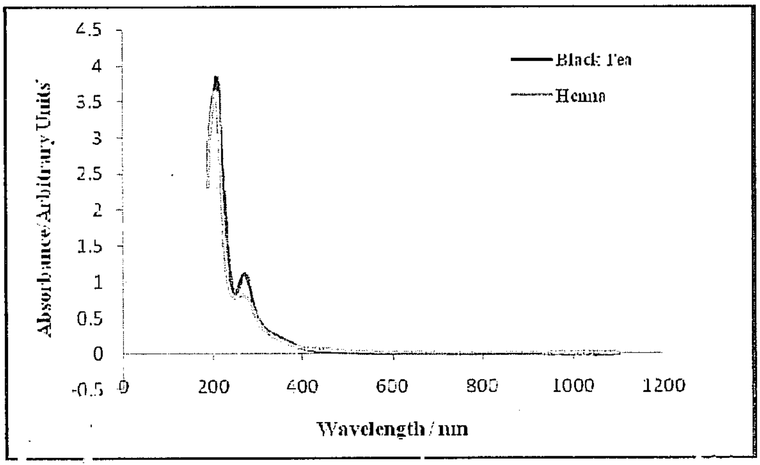

Figure 4. Absorption spectrum of pigments extracted from henna leaves and black tea

The open circuit voltage of ~371 mV and short circuit current of ~120 μA could be measured for cells sensitized with (henna + black tea) while it was ~ 233 mV and ~100 μA respectively for the cells sensitized only with pigments extracted from henna leaves at 100 $mWcm^{-2}$ light intensity. The increased photovoltage and photocurrent was attributed due to strong chelation of COOH group in the complex with TiO_2.

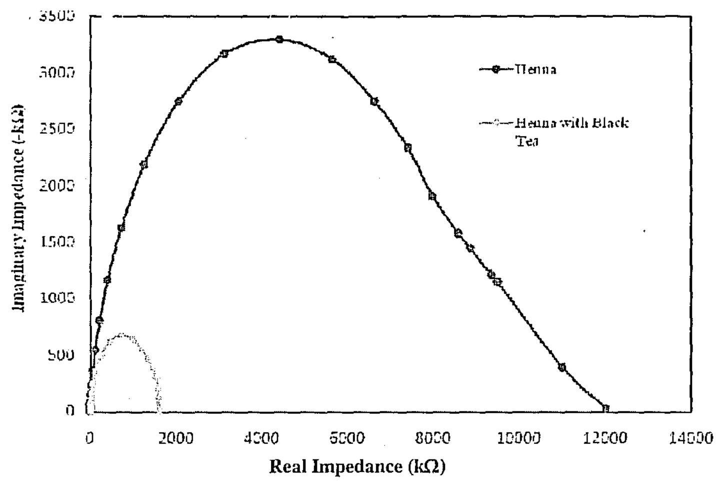

Figure 5. Nyquist plot of one of the samples made from henna and henna with black tea

The observed photocurrent is minute in both the cases as the light absorption occurs in the UV region. But when the chromophore of hennatannic acid bonds to amino acids like Theanine in this study, its colour become darken and showed increased photo electric properties due to its structural change. The impedance of TiO_2 film (10 μm) coated with henna was about 15.25 MΩ and it was decreased dramatically to 1.65 MΩ when the film is coated with (henna + Theanine) as seen in Figure 5. This observation supports that (henna + Theanine) complex gets attached to the TiO_2 film with the carboxylic group. That is because for the film only with henna, adsorption occurs with hydroxyl group attached to phenyl group which withdraws more electrons from the film increasing the impedance. But when the adsorption occurs with the carboxylic group in the (henna + Theanine) complex, it may enrich the electron concentration in the film which decreases the impedance. The main disadvantage of Theanine is that, it does not absorb any visible light. Therefore, experiments are underway to use amino acids which absorb visible light that will increase the efficiency of the DSSCs after chelating with henna.

CONCLUSIONS

In this study, we have succeeded to fabricate a DSSC with the complex of (henna + Theanine) which is extracted from tea. The photocurrent and photovoltage of this cell is greater than henna alone due to the stronger chealation of (henna + Theanine) complex and TiO_2 with the carboxylic group. The impedance spectroscopic measurements supported that the chealation of the complex with TiO_2 occurs with the carboxylic group. Several challenging problems remain to be resolved as this area is open for further studies of effective dye sensitizers in solar cells. However, the combination of Theanine in black tea with henna could serve as a model for the synthesis of other suitable pigments which motivates further research in DSSCs.

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