J. Myanmar Acad. Arts Sci. 2018 **Vol. XVI. No.2**

DYE-SENSITIZED SOLAR CELL BASED ON TiO_2-MgO COMPOSITE ELECTRODES BY USING COMBINED DYE

Shwe Yee Win[1], Moe Moe Aye[2], Than Than Win[3] and Yin Maung Maung[4]

Abstract

This research focused on improving the methodology of producing dye-sensitized solar cell (DSSCs) by the process of amalgamation with natural dyes. In this research, the *Sansevieria Trifasciata* dye as chlorophyll and *Gardenia* yellow dye as anthocyanian have been prepared and mixed these dyes. The combination of natural dyes by the ratio (1:1) used in the DSSCs as sensitizer. Next the titanium dioxide (TiO_2) and magnesium oxide (MgO) composite materials were mechanochemically prepared. And TiO_2 and MgO were mixed in different ratios as TiO_2 (pure), TiO_2 (95%)-MgO(5%) and TiO_2(90%)-MgO(10%). Then, the TiO_2 – MgO powder were prepared to be paste and they were deposited onto ITO/glass substrate by rolling method and immersed in the mixed dye solution to get dye cells. The dyed TiO_2-MgO-ITO/glass cells were offset with ITO/glass cell coated with adhesive carbon paste. The photovoltaic properties of prepared TiO_2-MgO DSSCs were investigated by J-V measurements. The results showed that the efficiency and fill factor (FF) were acceptable for industrial requirement and they are inexpensive candidates for DSSCs fabrication.

Keywords: *composite powders, ITO/glass, natural dye extract, carbon counter electrode*

Introduction

A solar cell which also known as photovoltaic cell is one of the promising options of renewable energy. Solar cell is divided into two groups which are the crystalline silicon and thin film. The dye-sensitized solar cells (DSSCs) which belong to the thin film groups, emerged as a new class of low cost energy conversion devices with simple manufacturing procedures (Mical G 2003). Dye-sensitized solar cell (DSSCs) is the third generation of solar cell which has been developed by O'Regan and Gratzel in 1991 (Suriati S et al 2015). The principal of operation of DSSCs is based on sensitization of a wide

1. PhD Candidate, Demonstrator, Department of Physics, Kyaukse University , Myanmar
2. Dr, Lecturer, Department of Physics, University of Yangon, Myanmar
3. Dr, Associate Professor, Department of Physics, Mandalay University of Distance Education, Myanmar
4. Dr, Associate Professor, Department of Physics, Mandalay University, Myanmar

band-gap metal oxide semiconductor to the visible light region by an adsorbed molecular dye (Lijian M et al 2015). In the fabrication of DSSCs, the choice material and the transparent electrode is crucial to obtain efficient light harvesting, charge separation and extraction. Improving the efficiency of the DSSCs can be achieved by coating a thin film of oxide layers on the TiO_2 electrode such as MgO, ZnO, Al_2O_3, Nb_2O_5, $CaCO_3$ and $SrTiO_3$ etc (Helin N et al 2014).

One of the probably best investigated photocatalysts is titanium dioxide (TiO_2). TiO_2 is a wide band gap oxide and it has been used as a photoelectrode in DSSCs. Because of its high specific surface area that allows the absorption of a large number of dye molecules (Kun C C et al 2014). TiO_2 has proved to be one of the most promising materials for various applications such as solar energy conversion, fuel cells, paints and photo catalysts, due to its high chemical stability, availability and low cost. Magnesium oxide (MgO) is another interesting material to catalyses oxidation reaction. Manganese compounds act as photocatalysts with oxygen as oxidative agent. For this reason, many attempts have to be performed to combine materials, titanium and manganese oxide, to get a composite material with advantageous catalytic activity (Boris M et al 2013). The sensitizers used in DSSCs were divided into two types: inorganic dye (includes metal complex, such as polypyridyl complexes of ruthenium and osmium) and organic dye (includes natural organic dyes and synthetically organic dye) according to the structure. Dye from the organic material is very interesting to develop because of the abundant natural (Suriati S et al 2015). Natural dye compounds generally contain anthocyanin, chlorophyll and carotenoids. The advantages of natural dyes are their low cost, easy extraction, non-toxicity and environmental benign nature. The energy conversion efficiency DSSCs is increased by adding a dye that absorbs light with wavelengths in the visible light range of the solar spectrum (Mounir A et al 2012).

Experimental Procedure

Preparation of TiO_2-MgO composite powders

Titanium dioxide (TiO_2) and magnesium oxide (MgO) were used in this work and they were prepared by using agate mortar, mesh-sieving and ball-milling method. Firstly, the TiO_2 was put in the agate mortar and it was ground for 3h to reduce the particle size. Then, the TiO_2 was applied by ball-milling method. The milling time interval was set for 3h. After ball-milling, the TiO_2 was also mesh-sieved with 3 step mesh to get uniform and the lightest particles. The ultrafine TiO_2 powder was heated by 800 ˚C for 1h. Finally, the undoped TiO_2 powder was obtained.

Next, the TiO_2 and MgO were prepared in different ratios. Firstly, the 95% of TiO_2 and 5% of MgO were mixed together. And they were prepared by using agate mortar, mesh-sieving and ball-milling method Finally, the homogeneous TiO_2 (95%)-MgO(5%) powder was obtained. The next one of 90% of TiO_2 and 10% of MgO were obtained in similar way. Finally, the homogeneous undoped TiO_2 and the different ratios of TiO_2-MgO composite powders were obtained.

TiO_2-MgO paste preparation

Firstly, 3 g of undoped TiO_2 powder was dissolved in 15 ml of acetic acid. During this preparation, detergent was added into it as a surfactant. Finally, the undoped TiO_2 paste was obtained. Similarity, TiO_2-MgO composite powders were prepared and TiO_2-MgO paste was formed. The undoped TiO_2 and TiO2-MgO paste were coated on conducting side of ITO glass using rolling method. In order to be dried and strengthen the undoped TiO_2 and TiO_2-MgO paste coating, the plates were calcined 450 ºC for 1h. Finally, the undoped TiO_2 and TiO_2-MgO films (active area = 1cm x 1cm) were formed on ITO/glass substrate.

Preparation of dye sensitizer

The anthocyanins pigments were extracted from *Gardenia yellow* and the chlorophyll were obtained from *Sansevieria Trifasciata*. First of all, *Gardenia yellow* was dried at room temperature for one week and they cut a small piece. After that, they were mixed with ethanol to eliminate the piquancy. After mixing, it was annealed at 80ºC for 1h with water-bath and extract solution was formed. After cooling, it was filled with filter paper and the pH level was 6.3 and the colour was found to be yellow. Secondly, *Sansevieria Trifasciata* was cut to become small pieces and they were mixed with ethanol. After mixing, it was annealed at 80ºC for 1h with water-bath and the dark green ethanol solution obtained from this procedure is filtered according to the same procedure previously. Then, the pH level was 6.25. After preparing these two dyes solution, they were combined with ratio (1:1) and the pH level was 6.3. And the combined dye solution was obtained. The process was shown in Fig 1.

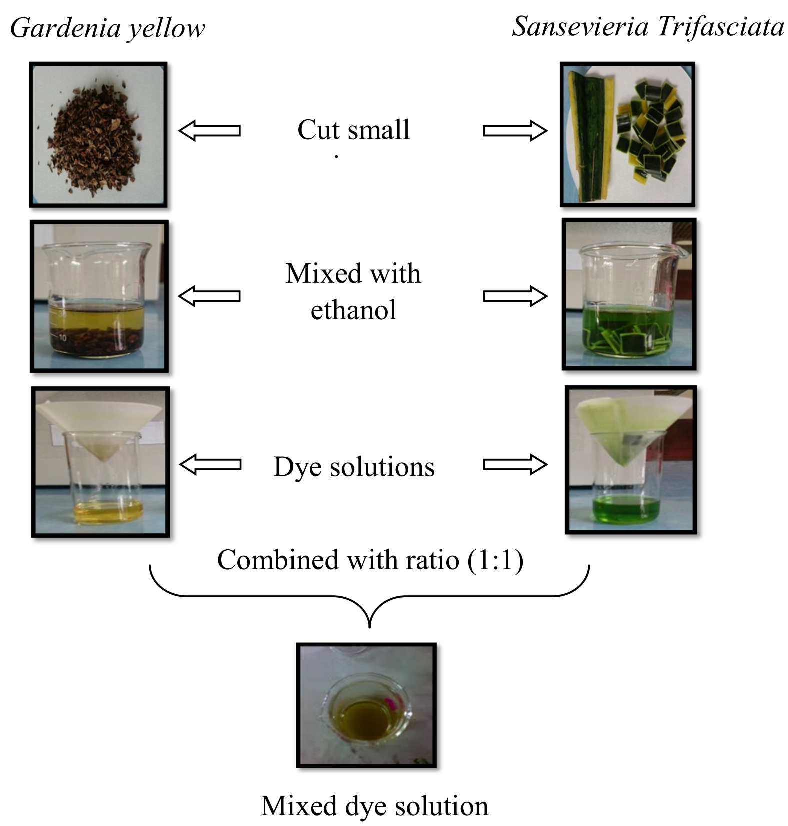


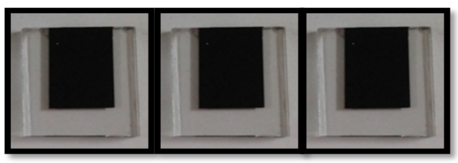
Figure 1. Process of dye solution preparation

Preparation of carbon catalyst

First of all, the KOH (16 ml) and ethanol (8 ml) were mixed together. Then, the carbon powder and the black carbon powder were dispersed into this mixture solution. After dispersion, carbonxylmethylcellulose (0.24 g) was also added and adhesive carbon paste was formed. It was coated onto ITO/glass substrate (active area = 1 cm x 1 cm) by rolling and annealed at 180ºC for 1 h.

Assembling of DSSCs

The undoped TiO_2 and TiO_2-MgO electrodes were immersed in mixed dye solution for 1 day and they were calcined at 200 ºC for 30 min. Fig 2 showed the carbon coated and TiO_2-MgO coated electrodes.



Carbon coated electrodes

TiO_2-MgO coated electrodes

Figure 2. Carbon coated and TiO_2-MgO coated electrodes

Preparation of the positive and negative electrodes were completed, 1-2 drops of mediator were placed on the negative electrode. Two prepared glass slides were set together and the sandwiching of the two plates were offset so that each one had a small position exposed so that an alligator clip could be attached and indicated in Fig3.

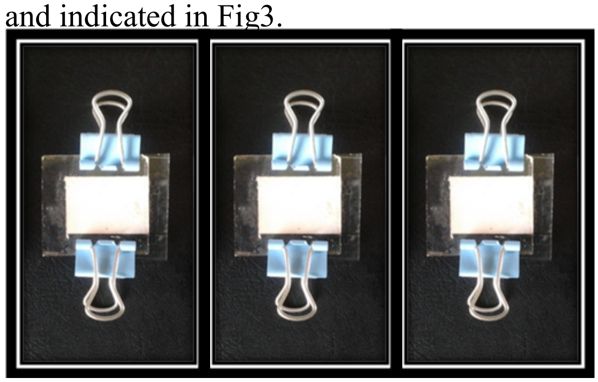


Figure 3. DSSCs with alligator clip

Results and Discussion

Characterization of TiO_2-MgO composite powder

The XRD analysis of the undoped TiO_2 and the different ratios of TiO_2-MgO composite powders were shown in Fig 4(a-c). All the peaks height and peak position were in good agreements with standard JCPDF library file. On the undoped TiO_2 XRD pattern, five peaks were clearly observed. The (101) reflection peak becomes more intense and sharper and the crystal structure of undoped TiO_2 was tetragonal. According to XRD analysis, the crystalline phase of the composite was tetragonal structure, depending mainly on the Mg content and it was found that MgO was influential as composite at this temperature.

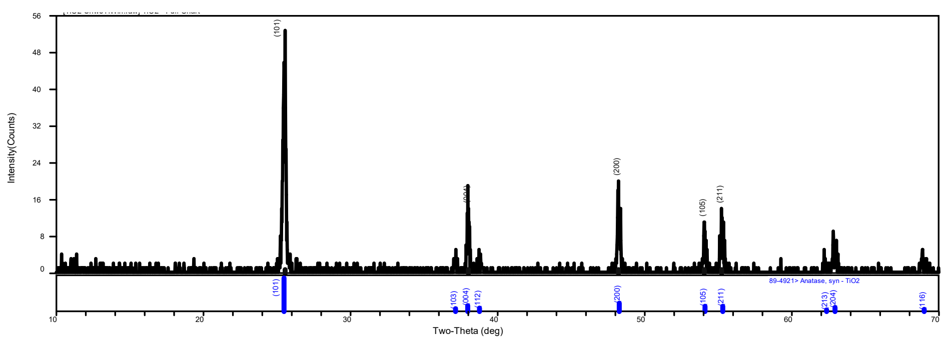


Figure 4(a). XRD patterns of undoped TiO_2 powder at 800˚C

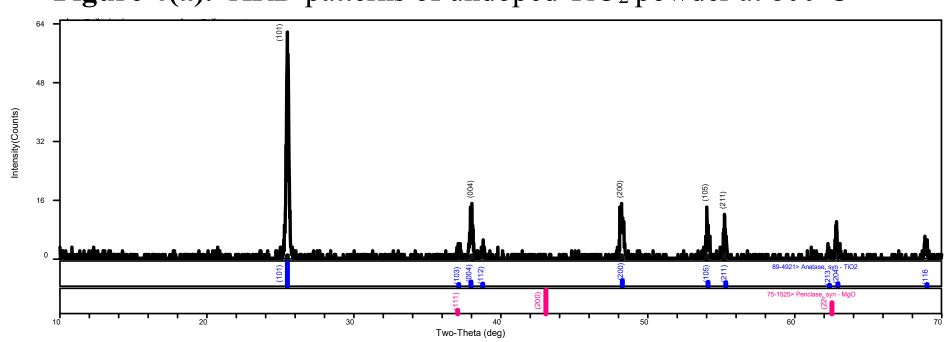


Figure 4(b.) XRD patterns of TiO_2(95%) –MgO(5%) at 800˚C

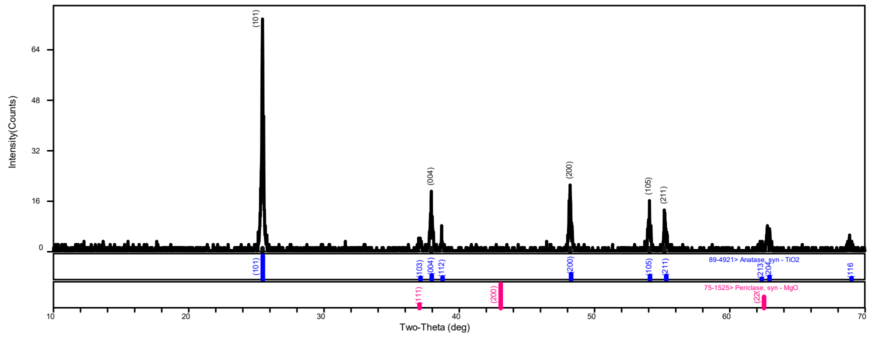
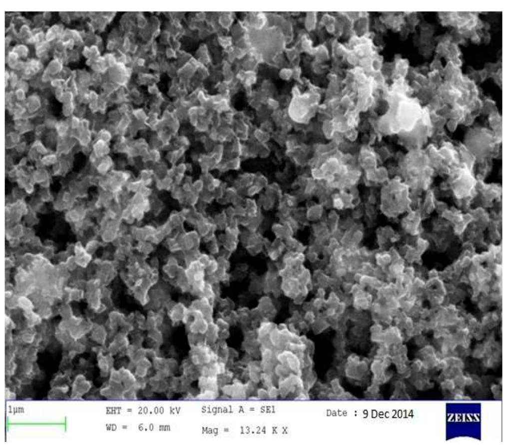


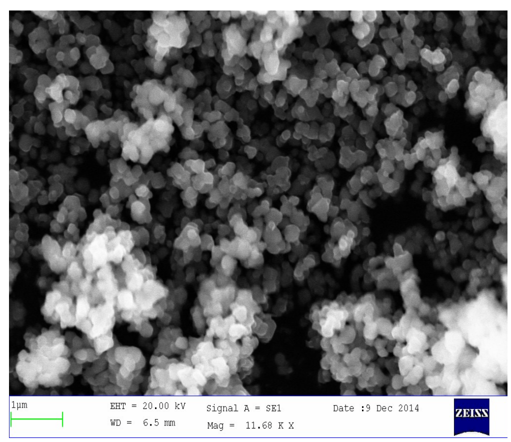
Figure 4(c). XRD patterns of TiO_2(90%)-MgO(10%) at 800˚C

Surface Morphology

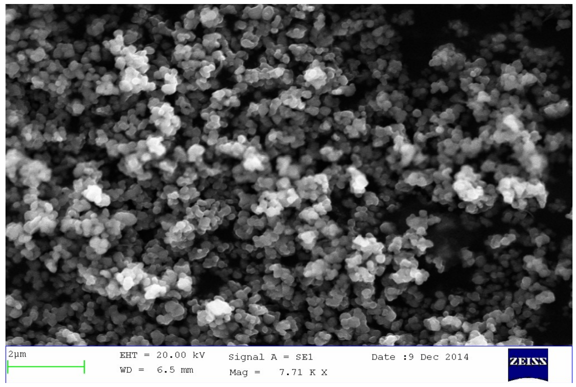
FESEM microphotograph of pure TiO_2 powder, TiO_2 (95%)-MgO(5%) powder and TiO_2(90%)-MgO(10%) powder were shown in Fig 5(a-c). Around the examined area, the grain size of undoped TiO_2 was estimated to be 0.233µm. And the grain size of TiO2 (90%)-MgO(5%) composite was estimated to be 0.215 µm and the TiO_2 (90%)-MgO(10%) composite was examined to be 0.135 µm. The grain size was significantly reduced with the increase of the Mg content. In addition, overall observation of TiO_2-MgO composite indicated a good microstructure with no discontinuities in terms of microracks. Apparently, with the increase of Mg content, this morphology was still observed, becoming denser and smoother.



(a) Undoped TiO_2



(b) TiO_2(95%)-MgO(5%)



(c) TiO_2(90%)-MgO(10%)

Figure 5. FESEM image

Optical Analysis

The absorption spectrum of mixed dyes was obtained from UV-Vis spectroscopy. The wavelength range of spectrum lay between 300 nm and 700 nm. . In this absorption spectrum, it was observed that the wide range absorption peaks and all of absorption peaks were within visible region. The energy band gap (E_g) for sample was 2.7526 eV. The UV-Vis absorbance spectra of mixed dye solution with ethanol were shown in Fig 6.

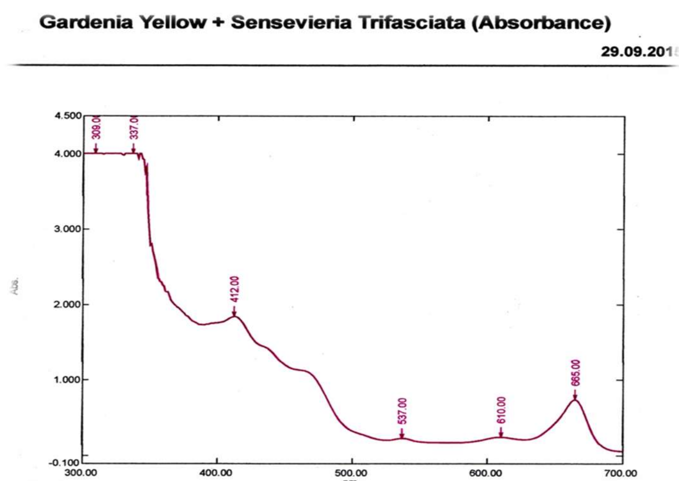


Figure 6. UV-Vis absorbance spectra of mixed dye solution with ethanol

Solar Cell Evolution

Fig 7(a-c) showed the change in photocurrent as a function of voltage with mixed dye solution. Photovoltaic cell parameters such as open-circuit voltage Voc, the short-circuit current density J_{sc}, the fill factor (FF) and conversion efficiency were depicted in Table 1.

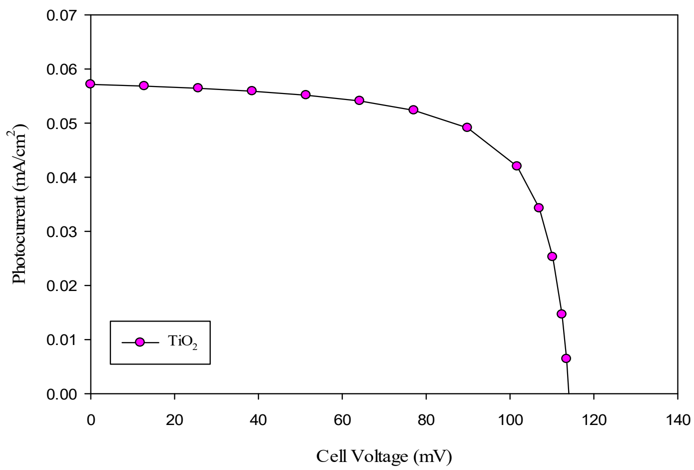


Figure 7(a). Current voltage characteristic curve of undoped TiO_2 cell

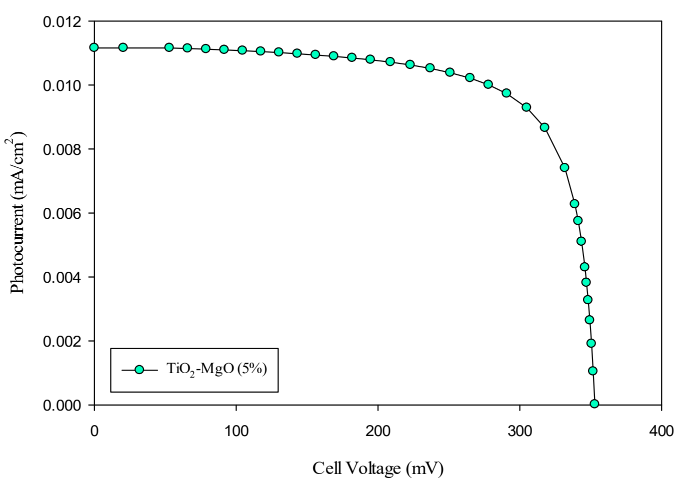


Figure 7(b). Current voltage characteristic curve of TiO_2(95%)-MgO(5%) cell

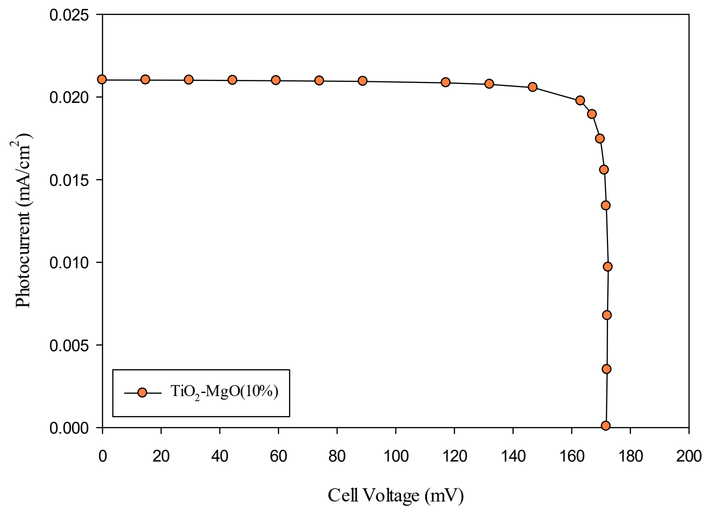


Figure 7(c). Current voltage characteristic curve of TiO_2 (90%)-MgO(10%) cell

Table 1. Photovoltaic parameters of all DSSCs cells with mixed natural dye sensitizer extract

Cell	J_{sc} ($mAcm^{-2}$)	V_{oc} (mV)	FF	η(%)
undoped TiO_2	0.048	114	0.67	0.348
TiO_2(95%)-MgO(5%)	0.009	178	0.72	0.236
TiO_2(90%)-MgO(10%)	0.029	172	0.88	0.259

Conclusion

Preparations of undoped TiO_2 and TiO_2-MgO composite electrode with combined dye extract have been implemented. The XRD studies confirmed that magnesium oxide coated titanium oxide fine powder was totally formed at given temperature. FESEM results indicated that the grain sizes for these powders were 0.233 μm, 0.215 μm and 0.135 μm, also tended to nano-sized grain. The optical absorbance spectrum for a combination of dyes (*Gardenia yellow* and *Sansevieria Trifasciata*) with mixing ratios (1:1) shows two absorption peaks in the visible region at the wavelength 537 nm (anthocyanine) and 610 nm (chlorophyll). Consequently, the combination of anthocyanine and chlorophyll gives a good absorption range for each of the red, green and blue wavelength. And the energy band gap (Eg) for a combined dye is 2.7526 eV. From I-V variation under illumination, it was significant that V_{OC}-J_{SC} (open-circuit voltage – short-circuit current) behavior was formed for all fabricated films. So, undoped TiO_2, TiO_2 (95%)-MgO (5%) and TiO_2 (90%)-MgO (10%) nanocomposite films exhibited the photovoltaic properties. As a result of photovoltaic evaluation, the undoped TiO_2 film possessed the best conversion efficiency as 0.348%. The fill factors of both DSSCs were found to be within the range of accepted value for industrial purposes. The research done is certainly said to be of low cost and by simply technique. Thus, the experimental data resulted from this research are quite promising, credible and applicable in use for DSSCs with combined dye sensitizer.

Acknowledgement

I would like to thank to Professor Dr Khin Myat Thu, Head of Department of Physics, Kyaukse University, for her kind permission to carry out this research. This research was totally done at Department of Physics in University of Yangon, Myanmar (2016-2017).

References

1. Boris. M., et al (2013) "Manganese/TiO_2 Composite Prepared and Used for Photocatalytic Active Textiles". CROATICA CHEMIC ACTA **86** 143

2. Helin. N., et al (2014) "Visible-Light Active and Magnetically Recyclable Nanocomposites for the Degradation of Organic Dye". Materials **7** 4034

3. Kun. C. C., et al (2014) " A Study of Mixed Vegetable Dyes with Different Extraction Concentrations of Use as a Sensitizer for Dye-Sensitized Solar Cells". International Journal of Photoenergy 1

4. Kartika. S. et al (2011) "Characterization of Optical Properties of the Sansevieria trifasciata Extract as Dye Sensitized Solar Cells (DSSC)". International Journal of Basic Applied Science IJBAS-IJENS **11** 10

5. Liberty. L. M et al (2012) "Effect of Calcination Temperature and MgO Crystallite Size on MgO/TiO_2 Catalyst System for Soybean Transesterification". World Academy of Science, Engineering and Technology **64** 889

6. Lijian. M., et al (2015) "Preparation and characterization of Due-sensitized TiO_2 nanorod solar cells". Thin Solid Films **557** 103

7. Mounir. A., et al (2012) "Studying of Natural Dyes Properties as Photo-Sensitizer for Dye Sensitized Solar Cells (DSSC)" Journal of Electron Devices **16** 1369

8. Mical. G. et al (2003) "Dye sensitized solar cells". Journal of Photochemistry C: Photochemistry Review **4** 145

9. Mark. L, et al (2014) "Analysis of energy gap opening in graphene oxide", 012003 (526)

10. Novoselov K.S, et al (2004) "Electric field effect in atomically thin carbon films" *Science* **306** 666–669

11. Suriati. S. et al (2015) "Materials for Enhanced Dye-Sensitized Solar Cell Performance: Electrochemical Application" International Journal of Electrochemical Science **10** 2859

12. Sungjin. P. et al (2009) "Chemical methods for the production of Graphene", Natural nanotechnology 132

13. Totao. Q, et al (2013) "Facllely prepared Polypyrrole reduced graphene oxide composite with crumpled surface for high performance supercapacitor electrode", ESI