Preparation and Investigation of ZnS-TiO$_2$ Thin Film Using Doctor-blade Method for Dye-sensitized Solar Cell

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Abstract

The ZnS-TiO$_2$ thin film for solar cells was prepared with a doctor-blade coating machine. A fluorine-doped tin oxide (FTO) was used as a substrate material. A sandwich design was built by 2.5 cm ×2.5 cm active area of the FTO glass substrate. The counter electrode layer substrate was coated with active carbon. The performance of dye-sensitized solar cells (DSSCs) based on chlorophyll and anthocyanin dye extracted from *Lagenaria siceraria* and *Tectona grandis* were analyzed in this paper. The current-voltage measurements of DSSCs were characterized under solar radiation with a digital source meter over the range from 0 V to 550 mV. The important parameters of DSSCs based on natural dye extracted from *Lagenaria siceraria* and *Tectona grandis*, such as the open-circuit voltage (V_{oc}), short circuit current (I_{sc}), fill factor (F_f), and electricity conversion efficiency (η)were determined from the current-voltage characterizations.

Keywords: *ZnS-TiO$_2$, Chlorophyll dye, Anthocyanin dye, digital source meter.*

1. INTRODUCTION

The solar energy technology is an important technology because the demand for energy is going up day by day. As well as, fossil fuels speedy resource consumption and environmental impurity have progressively become a worldwide interested since the past few decades [1]. A solar cell dependant on natural dye (DSSC) is observed as an inexpensive solar cell that includes in a thin-film solar cell [1,2,3]. For generally producing of DSSCs, these devices are based on a nanostructured metal oxide film and sensitized by an adsorbed dye molecule to capture the visible light [2].It is set up an oxide semiconductor device that is made up between a working electrode and an electrolyte; a photoelectric effect system. These cells are made up of three basic components: a dye sensitizer absorbing visible light to generate excitons and to transfer electrons from the excited state into the material conduction band, an electron-transporting layer, in which the interjected electrons travel throughout the electrode.The dye is again generated by an electron donor in the iodine solution [1]. In developing solar technology, silicon solar cells have not got demand due to their high cost and the modern tech of silicon-based raw material production [1,2]. At a recent time, the photovoltaic cells based on organic dye sensitizer have been investigated as the third generation of photovoltaic (solar) cells that it can transform any visible light into electrical energy. So, the researchers interested in the properties of DSSCs such as environmental friendliness, flexibility, low cost, and abundant materials, as well as high efficiencies even below cloudy and artificial light sources [1,2,3,4]. This cell was invented by Michael Grtzel and Brian O' Regan at the Ecole Polytechnique Federale de Lausanne in 1991 and so it is also known as Grtzel cells. For his invention, Michael Grtzel was awarded the 2010 Millennium Technology Prize [2,3,4-6].

Several natural dyes, such as anthocyanin, chlorophyll, tannin, and carotene, extracts from various plants, fruits, flowers, and leaves, and bulbs have been applied in solar cells to be the natural dye sensitizers [6]. In my research, the extracted dyes from gourd leaves and teak leaves were be used in DSSC. Moreover, green pigment has been used in dyeing food, drink, soap, cosmetics, and cloth. Because of one of the properties, stability, these pigments have been practiced for coating materials and anti-knocking agents in gasoline engines. It is also discovered that the *Lagenaria siceraria* plant is one of the green plants containing chlorophyll [7]. In many research works, the natural organic dye was extracted from several parts of the plant by the use of various types of organic solvents. Organic solvents were found to regard the absorption of the dye and the binding between the dye and semiconductor surface. Apart from the properties, anthocyanin regarded as health beneficial compounds can advance scientific interest in these pigments. Naturally, plants reveal characteristic colors because they have strong pigments [8]. Further, teak leaves (*Tectona grandis*) can be utilized as a natural dye.

Since they contain anthocyanin, a source of brown color [9]. Natural dyes contain carboxyl, hydroxyl and so on. They can easily react with the surface of nano TiO_2. During the recent decades, people have conducted extensive research in the naturally dye-sensitized DSSCs. Firming the growing of the economy, environmental pollution and energy shortage problems have become more and more serious. Hence, the natural organic dye in DSSC was used in solar energy, which is highly efficient, and environmentally friendly cleaning [9,10,11].

Zinc sulfite (ZnS) is an II-VI compound semiconductor with a direct and broad bandgap of 3.68 eV at room temperature and applied as a phosphor in the optical devices [12,13,14-17]. The photoelectrical properties create ZnS particles as a very promising element in solar cell application [17]. The interaction between the zinc sulphite and titania phases and the strong adsorption to the substrate at the composite surface are responsible for the raise photocatalytic activity [18]. Titanium dioxides are got from a variety of ores. They have tetragonal and orthorhombic structures. They have been widely utilized as photocatalyst in the abasement of organic dyes because of the stability of their chemical structure, non-toxicity, optical and electrical properties [18]. Likewise, it is examined that the synthesis of ZnS coupled with semiconductor TiO_2 induces the optical response towards the visible region [18,19-21].

2 EXPERIMENTAL PROCEDURE

2.1 Preparation of ZnS-TiO_2 composite

Zinc sulfite (ZnS) and titanium dioxide (TiO_2) powder were used as starting chemicals. ZnS and TiO_2 were mixed with methanol by solid-state mixed oxide route by equal molar ratio. The milling process was performed to form the homogeneous and small particle size. The ball milling process was performed for 5 h. After ball milling, ZnS-TiO_2 was annealed at 500ºC for 1 h in the O_2 ambient. And then it was ground to a fine powder by using agate motor for about 9 h. It was sieved by 3-stages mesh (100, 150, 200) and the uniform particle-sized powder was formed.

2.2 Preparation of FTO glass

2.5 cm × 2.5 cm fluorine-doped tin oxide (FTO) glass pieces for the electrodes were first cleaned using soapy water for 5 minutes, then rinsed with deionized water for 15 minutes and cleaned again with 2-propanol for 15 minutes and dried in an oven at 105ºC for 30 minutes. They were already prepared to use in DSSC.

2.3 Preparation of working electrode and counter electrode with FTO glass

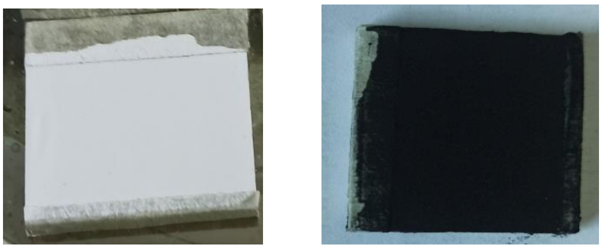
Working electrode was prepared from particles of ZnS-TiO_2 composite. Earlier being deposited, FTO glass resistance was tested by using a digital multimeter to determine the conductive side. The counter electrode was prepared from carbon powder and 2methoxylethanol. The carbon counter electrode was heated in an electric furnace at 160ºC for 40 minutes. Working electrode was prepared from 1 g of ZnS- TiO_2 composite powder and 10 mL of 2methoxylethanol deposited onto the conductive glass surface. The working electrodes were conducted at the temperature of 600ºC for 30 minutes in the furnace. The ZnS-TiO2 solution for the working electrode and carbon solution for counter electrode were deposited onto the conductive surface of the FTO substrate by using the doctor-blade coating machine. Working electrode and counter electrode were banked on conducting the surface of FTO glass with a dimension of 2.5 cm× 2.5 cm.Figure.1 (a), and (b) were working electrode and counter electrode. Figure.2. was the home-made doctor blade coating machine.

2.4 Preparation of natural dyes

Gourd leaves (*Lagenaria siceraria*) and teak leaves (*Tectona grandis*) were firstly collected to extract the natural dyes. After that, fresh water was used to clean them and they were dried at room temperature for about one week. These dried leaves were made into small pieces. The natural dyes from these dried leaves were extracted with ethanol solvent by using a soxhlet extractor. For adsorption on ZnS-TiO_2, the FTO/ZnS-TiO_2 slides were individually equilibrated in the respective dye solution for 2 hours after which the glass slide was immediately cleaned with excess ethanol solvent and dried in gentle nitrogen flow to remove any water content. The soxhlet extractor and the extracted dyes were shown in Figure.3 (a), (b), (c), and (d).

2.5 Assembling of the complete DSSCs

The working electrode and the counter electrode were overlapping placed in a holder so that the ZnS-TiO_2 composite area of the working electrode was the only part of it that was in contact with the counter electrode. The non-composite layer area of the working electrode and the non-carbon layer of the counter electrode were attached to the measuring equipment using cords and binder clips. The electrolyte solution was pipette dropped between the electrodes. The complete DSSCs were shown in Figure.4. Under solar radiation, the current and voltage (I-V) characteristics for solar cells based on gourd dye, teak dye, and mixed dye were measured with a Digital Source Meter (Model 2450, Keithley).



(a) (b)

Fig.1. (a) Working electrode using ZnS-TiO_2 composite, and (b) Counter electrode using carbon

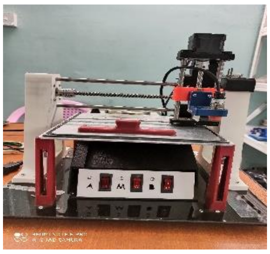


Fig.2. Doctor-blade coating machine

(a) (b) (c) (d)

Figure.3.(a) Soxhlet extractor, (b) gourd dye solution, (c) teak dye solution, and (d) mixed dye solution

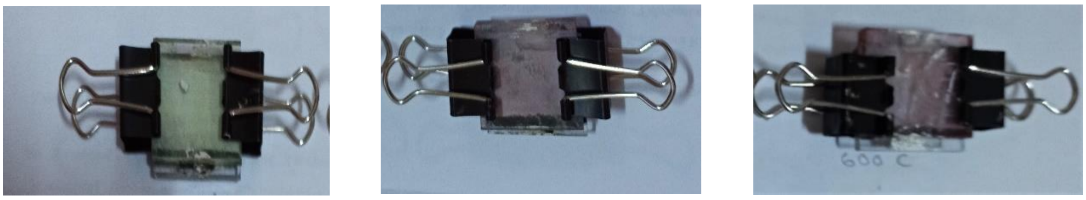


Fig.4. Pairs of electrodes with the doctor-blade coating method for gourd, teak, and mixed leaves.

3 Results and Discussion

3.1 Current-voltage characteristics of dye-sensitized solar cells

The solar cell parameters of natural dye-sensitized solar cells based on chlorophyll and anthocyanin dye extracted from *Lagenaria siceraria* and *Tectona grandis* were analysed by measuring the current and voltage (I-V) characteristics using source meter under solar radiation. The important parameters of DSSCs based on natural dye extracted from *Lagenaria siceraria* and *Tectona grandis*, such as the open-circuit voltage (V_{oc}), short circuit current (I_{sc}), fill factor (F_f), and efficiency (η) were assessed from the current-voltage characterizations. The current-voltage curves of the DSSCs using the natural dye sensitizers extracted from gourd, teak, and mixed dyes were shown in Figure.6. (a), (b), and (c). Table 1 showed the important parameters of the DSSC related to the gourd, teak, and mixed sensitizers. Fill factors (F_f) for solar cells based on gourd, teak, and mixed leaves were determined by the following equation,

$$F_f = \frac{P_{max}}{V_{oc}\, I_{sc}} = \frac{I_{max}V_{max}}{V_{oc}\, I_{sc}}$$

The fill factors of extracted dyes were calculated to be 0.075 for gourd, 0.08 for teak, and 0.078 for mixed leaves, respectively. There was found that these results were less differences.

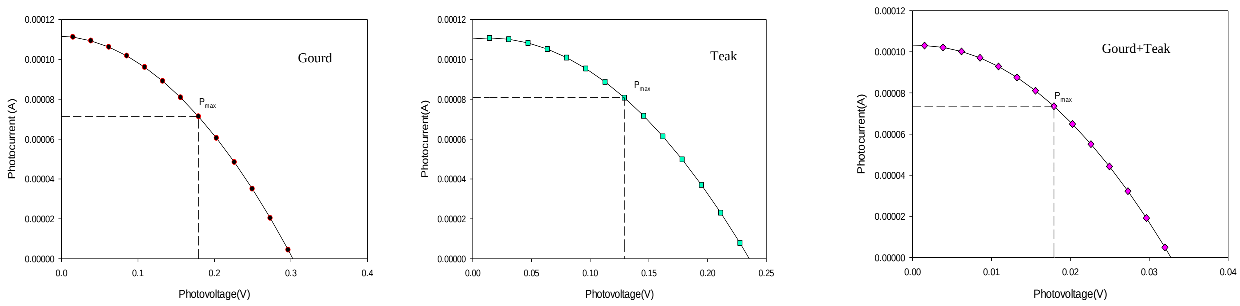
Then, the conversion efficiency (η) of the DSSCs with electrical power under solar radiation was resolved by the following equation,

$$\eta = \frac{P_{out}}{P_{in}} = \frac{I_{sc}V_{oc}F_f}{P_{in}}$$

Where, P_{in} is incident optical power on the solar cells. The power conversion efficiencies were determined to be 2.30 % for gourd, 1.87 % for teak, and 0.24 % for mixed leaves. From the results, the maximum efficiency was found to be 2.30 % for gourd leaves, and the minimum was calculated to be about 0.24 % for mixed leaves.

Table 1. Short circuit current I_{sc}, open-circuit voltage V_{oc}, fill factor F_f, and power conversion efficiency η for Dye-sensitized solar cells (DSSCs)

Sensitizers	J_{max} (mA/cm^2)	V_{max}(V)	J_{sc}(mA/cm^2)	V_{oc}(V)	F_f	P_{in} (mW/cm^2)	η (%)
Gourd	0.0143	0.1793	0.112	0.303	0.075	0.1113024	2.30
Teak	0.0162	0.1291	0.111	0.235	0.080	0.1113024	1.87
Mixed	0.0147	0.0179	0.104	0.033	0.078	0.1113024	0.24



(a) (b) (c)

Fig.6. Current-voltage curves of the DSSCs using the sensitizers extracted from (a) gourd (*Lagenaria Siceraria*) leaves, (b) teak (*Tectona Grandis*) leaves, and (c) mixed leaves

4 Conclusions

In this research paper, the DSSCs related to natural dye sensitizers have been implemented. The solar cell parameters were determined from I-V characteristic curves. From the results, the maximum conversion efficiency was decided to be about 2.30% from *Lagenaria Siceraria* based cell. The minimum was found to be 0.24% from mixed dye cell, and the efficiency of *Tectona Grandis* was obtained about 1.87%. The conversion efficiency of *Lagenaria Siceraria* dye cell was found to be the best of three natural sensitizers. One of the reasons for this could be the higher amount of *Lagenaria Siceraria* dye adsorbed by the ZnS-TiO_2 thin film. And it is due to a higher intensity and broader range of the light absorption of the extract, the strong interaction between ZnS-TiO_2 and chlorophyll in the *Lagenaria Siceraria* extract and it leads to being a better charge transfer. Therefore, the natural dye extracted from *gourd leaves (Lagenaria Siceraria*) can be used to an alternative chlorophyll source for natural dye-sensitized solar cells.

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