

## PREPARATION OF LOW TEMPERATURE TiO<sub>2</sub> PHOTOELECTRODE FOR FLEXIBLE DYE SOLAR CELLS APPLICATION

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Diterima: 7 Agustus 2012

Diperbaiki: 7 Januari 2013

Disetujui: 14 Februari 2013

### ABSTRACT

**PREPARATION OF LOW TEMPERATURE TiO<sub>2</sub> PHOTOELECTRODE FOR FLEXIBLE DYE SOLAR CELLS APPLICATION.** Flexible dye solar cells were dye cell that were assembled using plastic transparent electrodes as substrate, so the cell processing must be applied in low temperature. In this paper, investigation of TiO<sub>2</sub> photoelectrode preparation will be described. We used a low temperature TiO<sub>2</sub> paste which deposited onto Indium Tin Oxide-PolyEthylene Terephthalate (ITO-PET) substrate using doctor blade printing technique. The annealing temperature of the TiO<sub>2</sub> layer was 120 °C, 130 °C and 140 °C for 4 hours. The optical properties were investigated by Ultra Violet-Visible Spectrophotometer. The performance of the cell was measured using a solar simulator with light at an intensity of 50 mW/cm<sup>2</sup> generated by a xenon lamp. An efficiency of 0.019 % was achieved for such cells with a 2.0 cm<sup>2</sup> active area under TiO<sub>2</sub> annealed in 120°C.

**Keywords:** Flexible dye solar cells, TiO<sub>2</sub> photoelectrode, Low temperature

### ABSTRAK

**PREPARASI FOTOELEK TRODA TiO<sub>2</sub> UNTUK APLIKASI SEL SURYA FLESIBEL.** Sel surya fleksibel berbasis pewarna merupakan sel surya yang dibentuk menggunakan elektroda transparan sebagai substrat, sehingga proses berlangsung pada suhu rendah. Pada tulisan ini diuraikan penelitian tentang preparasi fotoelektroda TiO<sub>2</sub> untuk sel surya fleksibel. Pada percobaan menggunakan pasta TiO<sub>2</sub> suhu rendah yang dideposisi pada permukaan substrat *Indium Tin Oxide-PolyEthylene Terephthalate (ITO-PET)* dengan teknik *doctor blade*. Suhu *annealing* lapisan TiO<sub>2</sub> divariasikan pada 120 °C, 130 °C and 140 °C selama 4 jam. Sifat optik dianalisis menggunakan *UV-Vis Spectrophotometer*. Kinerja sel diukur menggunakan Solar Simulator lampu Xenon, dengan intensitas cahaya 50 mW/cm<sup>2</sup>. Efisiensi sel terbaik sebesar 0,019 % dicapai sel dengan *area* aktif 2 cm<sup>2</sup> yang menggunakan fotoelektroda TiO<sub>2</sub> yang *dannealing* pada suhu 120 °C

**Kata kunci:** Sel surya fleksibel berbasis pewarna, Fotoelektroda TiO<sub>2</sub>, Suhu rendah

### INTRODUCTION

Since Dye Sensitized Solar Cell (DSSC) was reported firstly researcher, the solar cells research has become attention of scientist and academics due to its low cost and simple preparation compared with conventional silicon solar cells. Beside the high conversion efficiency, DSSC is also one of environmental friendly energy. The conversion efficiency of glass-based DSSC has been achieved about of 10-11%. [1,2]

Generally, the DSSC composed of a dye sensitized porous TiO<sub>2</sub> photoanode, I<sub>3</sub><sup>-</sup>/I<sup>-</sup> redox couple electrolyte, and a platinized counter electrode (Figure 1). In standard

DSSC, the porous titania films and Pt film are mounted on rigid transparent conductive oxide glass substrates. Transparent conducting glass is usually employed as substrate material of the DSC due to its excellent optical and electrical properties, and compatibility with high-temperature processing [3].

Recently, a novel type of DSSC employing flexible plastic conductive oxide as substrate has been developed. Unlike the glass-based DSSC, flexible DSSC can be used for application of mobile power for portable electronic devices due to the property of light weight,

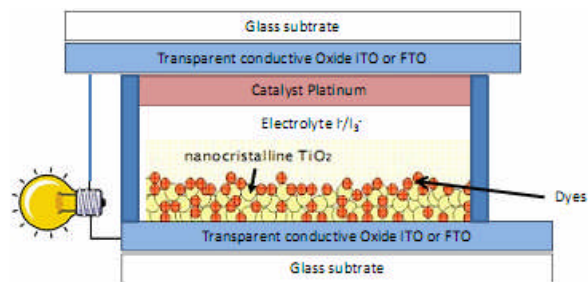


Figure 1. Device structure of DSSC [6]

thin and good flexibility. Flexible substrate which commonly used for the deposition of  $\text{TiO}_2$  is a polymer of PolyEthylene Terephthalate (PET) or PolyEthylene Naphthalate (PEN) which coated with transparent conductive oxide of ITO (Indium Tin Oxide). Substrate such plastic foils have been used to fabricate flexible DSSC using roll-to-roll production techniques that can be applied to cost-effective mass [4,5].

Several alternative approaches to the deposition  $\text{TiO}_2$  that can be used, are *doctor blade printing* [5,7], *spray deposition* [8] and *electrophoretic deposition* [9]. One of the problems of plastic substrate DSSC is has a lower efficiency than glass substrate DSSC. Temperature process annealing of nano porous  $\text{TiO}_2$  is limited below  $150^\circ\text{C}$ . It impacted on the low cell efficiency due to the  $\text{TiO}_2$  annealing process lasted less perfect that caused poor interparticle connectivity among the  $\text{TiO}_2$  particles [8].

In this paper, we reported the preparation of low temperature  $\text{TiO}_2$  photoelectrode using a suspension of  $\text{TiO}_2$  nanoparticles without any organic additive. The paste was deposited onto Indium Tin Oxide-PolyEthylene Terephthalate (ITO-PET) substrate by blade printing technique. The annealing temperature of the  $\text{TiO}_2$  layer was  $120^\circ\text{C}$ ,  $130^\circ\text{C}$  and  $140^\circ\text{C}$  for 4 hours. Our research aims to obtain effective temperature of annealing and to study its influence on optical properties. The photovoltaic performance was measured using a simulated solar light at an intensity of  $50\text{ mW/cm}^2$  generated by a xenon lamp.

## EXPERIMENTAL METHODS

### Preparation and Characterization of The $\text{TiO}_2$ Photoelectrode

The transparent Indium Tin Oxide coated PolyEthylene Terephthalate (ITO-PET) used for the conducting flexible substrate was purchased from Aldrich with sheet resistivity of  $60\text{ ohm/sq}$ . The substrate was washed with deionized water and isopropyl alcohol using ultrasonic cleaner.  $\text{TiO}_2$  paste which purchased from Solaronix (Nanoxide DL-series) was deposited onto substrate by doctor blade printing technique with a glass rod and scotch tape 3M as a frame and spacer. The  $\text{TiO}_2$

film was annealed in a vacuum oven at  $120^\circ\text{C}$ ,  $130^\circ\text{C}$ , and  $140^\circ\text{C}$  for 4 hours, respectively.

## Cells Fabrication

The  $\text{TiO}_2$  porous electrodes were immersed in ethanol solution of Ruthenium complex dye  $\text{RuL}_2(\text{NCS})_2 \cdot 2\text{TBA}$  ( $\text{L} = 2,2'$ -bipyridil-4,4'-dicarboxylic acid;  $\text{TBA} = \text{tetrabutylammonium}$ ) [10] known as N719 (Dyesol) for 24 hours at room temperature. Counter electrode were prepared by depositing a Pt onto PET substrate using sputtering technique. The reason for choosing sputtered-Pt as the catalyst has been explained in our previous work [11]. Deposition process was performed using Direct Current-sputtering process with an initial pressure of  $6.6 \times 10^{-3}\text{ Pa}$ , argon gas pressure of  $5.3 \times 10^{-1}\text{ Pa}$ , rotation speed of 5 rpm and power of 5 W, for 20 minutes.

Both of photoelectrode and counter-electrode were assembled into a sandwich structure using thermoplastic sealant  $60\text{ }\mu\text{m}$  thick (Solaronix S.A) as spacer. Finally, Dyesol liquid electrolyte EL-HSE was injected into the assembled samples and the remaining holes were subsequently sealed using a silicon rubber.

## Characterization

X-Ray Diffraction graphic were used to study crystallization of  $\text{TiO}_2$  film with varying the annealing temperature. The optical properties were investigated using Ultra Violet-Visible (UV-Vis) Spectrophotometer. The surface morphologies of the  $\text{TiO}_2$  film were observed using a Jeol Scanning Electron Microscope (SEM). Cell performances were measured using a National Instrument I-V measurement with  $50\text{ mW/cm}^2$  Xenon light source through an AM 1.5 filter. Incident Photon-to-current Conversion Efficiency (IPCE) was measured as an action of spectrum applying IPCE measurement apparatus.

## RESULTS AND DISCUSSION

Figure 2 shows the X-Ray Diffraction (XRD) patterns of the  $\text{TiO}_2$  film which based on variation of annealing temperature. Those patterns indicate  $\text{TiO}_2$

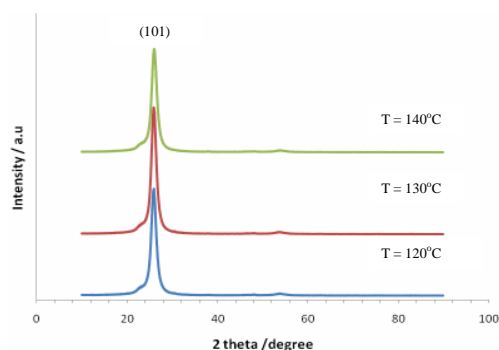


Figure 2. XRD patterns of  $\text{TiO}_2$  film based on variation of annealing temperature

crystallites are pure anatase phase. The average crystallite size was calculated according to Scherrer's equation, approximately 11 nm (at  $2\theta = 25.93^\circ$ ), 41 nm (at  $2\theta = 25.66^\circ$ ) and 12 nm (at  $2\theta = 25.98^\circ$ ), for annealing temperature of 120 °C, 130 °C and 140 °C, respectively. The DSSC performance somewhat depends on the size and crystallinity of TiO<sub>2</sub> particle. Wang et.al [12] reported that the higher photoconversion efficiency could be achieved by smaller TiO<sub>2</sub> particles due to the effectiveness in dye absorption

Based on XRD analyze, nanoporous TiO<sub>2</sub> which is thermal annealed at 120 °C has smaller crystallite size. The scanning electron microscopy image of surface TiO<sub>2</sub> film which coated onto Indium Tin Oxide coated PolyEthylene Terephthalate (ITO-PET) substrate by blade coating technique after annealed at 120 °C for 4 h is shown Figure 3(b). The film has TiO<sub>2</sub> nanoparticle surface homogenously with thickness of 1µm. Figure 3(a) and Figure 3(b) indicate that low temperature can change structure TiO<sub>2</sub> film. In the Figure 3(b) demonstrates the formation of porous TiO<sub>2</sub> layer having a moderate porosity and high surface area to

where dye molecules could be adsorbed. It differs with Scanning Electron Microscope (SEM) image of layer deposited from the suspension of TiO<sub>2</sub> prepared without annealing treatment Figure3(a). As it is shown in Figure 3(b) there was TiO<sub>2</sub> particle agglomerates formed. It indicates that excellent inter particle connectivity for layer with annealing treatment.

Optical properties of film TiO<sub>2</sub> nanoparticle after annealed, with and without dye coated were indicated in Figure 3. The optical absorbance was measured in the visible region of 400 nm-800 nm to study the capability in photo-sensitizing the TiO<sub>2</sub> nanoparticles. In the figures show that TiO<sub>2</sub> film without dye which is annealed at 140 °C can absorb visible light in wider wavelength range than the others. It may be caused by the crystallinity of TiO<sub>2</sub> nanoparticle which is better at annealing temperature of 140 °C. According to the image, it can also be known that the addition of the dye will enhance the ability of TiO<sub>2</sub> electrode layer to absorb incoming light. The dye coated TiO<sub>2</sub> film which is annealed of 130 °C has good absorbance of visible light.

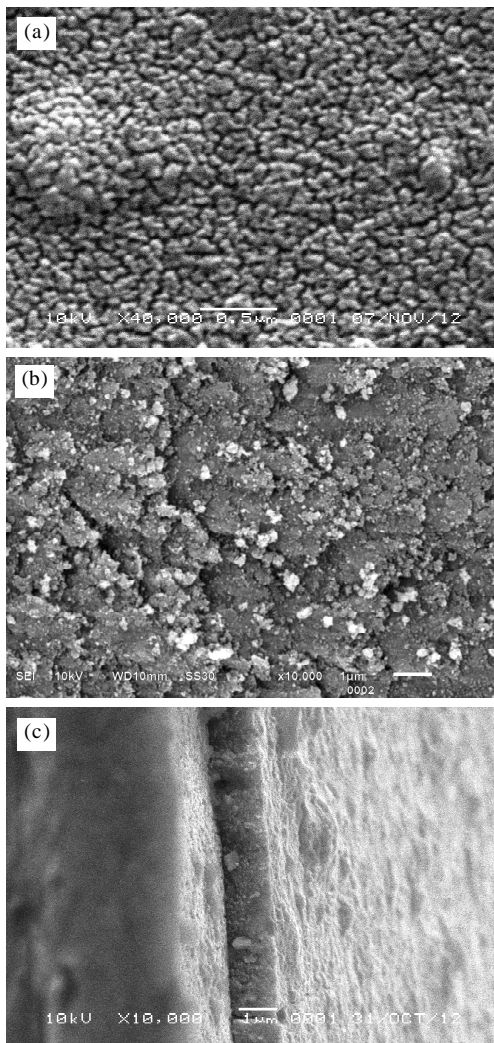


Figure 3. SEM image of TiO<sub>2</sub> film (a). before annealed, (b). annealed of 120 °C and (c). thickness

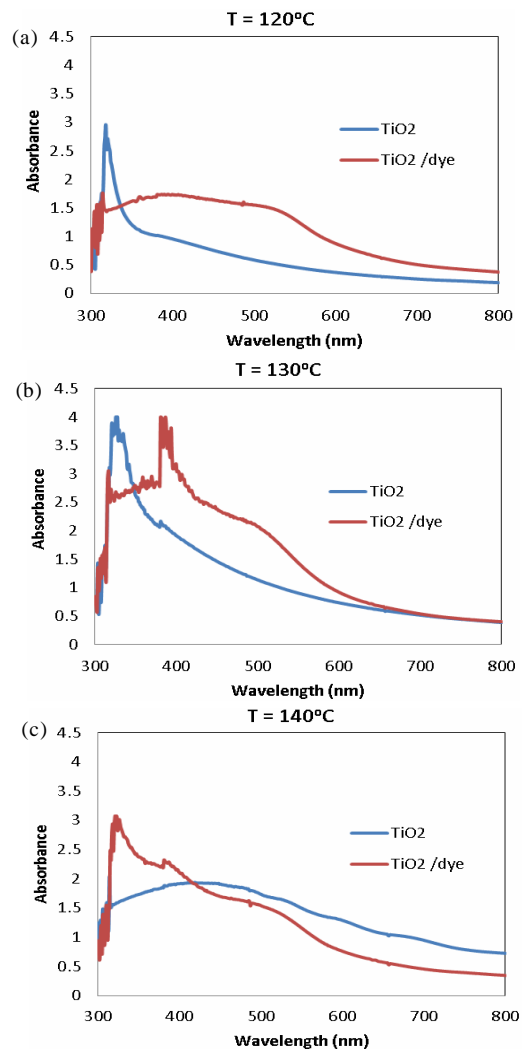


Figure 3. Optical properties photoelectrode film with dye (TiO<sub>2</sub>/dye) and without dye (TiO<sub>2</sub>)

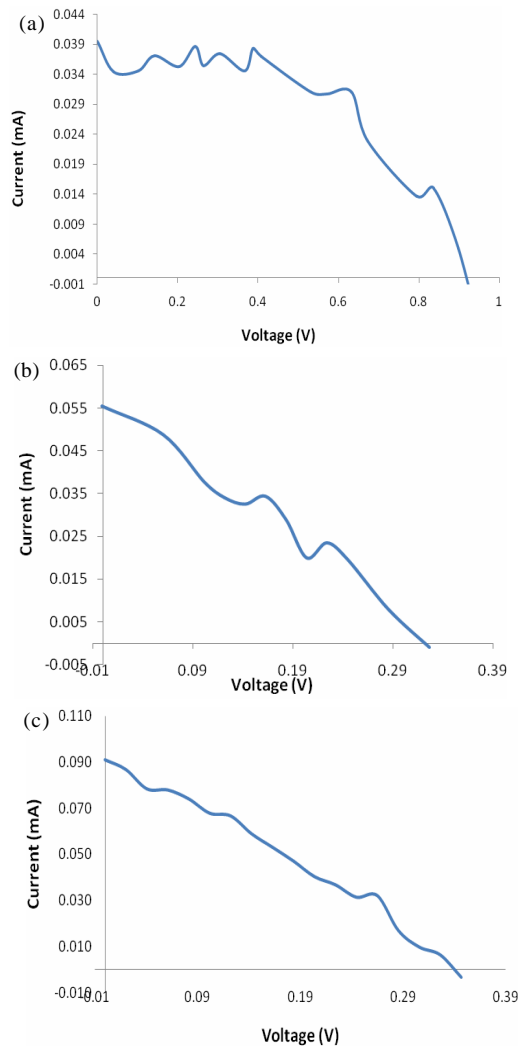


Figure 4. Characteristics I-V of DSSC : (a). 120°C, (b). 130°C and (c). 140°C

Table 1. Characteristics performance for fabricated cell

Characteristics	T = 120 °C	T = 130 °C	T = 140 °C
Open circuit voltage $V_{oc}$ (Volts)	0.938	0.306	0.326
Short circuit current $I_{sc}$ (mA)	3.94E-02	5.92E-02	8.69E-02
Maximum power $P_m$ (Watt)	1.96E-05	5.59E-06	8.65E-06
Fill factor FF	0.53	0.31	0.3
Efficiency (%)	0.019	0.0056	0.0087

The I-V characteristics for the cells with variation of annealing temperature are shown in Figure 4. The values of  $V_{oc}$ ,  $I_{sc}$ , FF and cell efficiency ( $\eta$ ) for fabricated cell using with N719 dye and Pt sputtered counter electrode of active area  $2 \text{ cm}^2$  illuminated by a Xenon lamp with an incident light of  $50 \text{ mW/cm}^2$  are summarized in Table 1. As shown in Table 1, generally the electric performances for fabricated cell are still low. In this investigation, it is found that the cell with  $\text{TiO}_2$  annealing of  $120 \text{ }^\circ\text{C}$  has a high open circuit voltage, as well as 0.938 V. The high values of  $V_{oc}$  could be attributed to the high density of

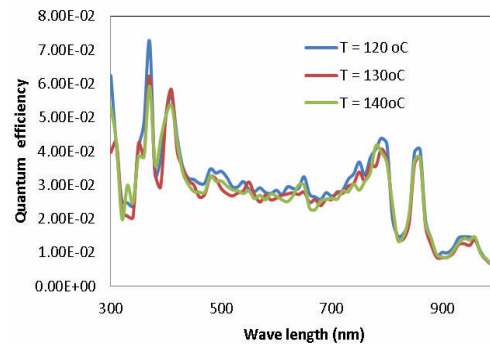


Figure 5. The IPCE spectra of cells with variation of temperature annealing

the  $\text{TiO}_2$  nano particles in the layer formed although the annealing treatment is in low temperature [6]. The contacts between nano particles  $\text{TiO}_2$  will improve the electron transfer from  $\text{TiO}_2$  electrode interface to the ITO-PET substrate.

The low value of short circuit current,  $I_{sc}$  is related to low external quantum efficiency which is limited by the optical absorption of the active layer. There is a correlation between the active layer thickness and the short circuit current resulted. The thickness relates on the total active surface area and the ability of the electrode in dye collection. In this study, the thickness  $\text{TiO}_2$  film which measured according to the SEM cross section Figure 3(c) is very thin thus just little dye sensitized that can be absorbed little.

The reported that there is a proportional relationship of the  $\text{TiO}_2$  film thickness and the conversion efficiency. The photocurrent of solar cells is proportional to the intensity of incident light; therefore, the amount of light transmitted by the substrate is related directly to the performance of a solar cell. Figure 5 shows the Incident Photon-to-current Conversion Efficiency (IPCE) spectra for cells. Generally, the cells have high absorption coefficient in the visible region of the spectrum 400 nm and 780 nm. The highest quantum efficiencies of each cells in the spectrum of 400 nm are around 0.06-0.08. In summary, in this investigation, the high quantum efficiency (IPCE) and the best performance of cells was resulted on the thermal annealing of  $120 \text{ }^\circ\text{C}$  for 4 hours. The conversion efficiency of 0.019 % was obtained with a short circuit photocurrent of  $3.94 \times 10^{-2} \text{ mA}$ , an open circuit voltage of 0.938 Volt, and a fill factor of 0.53.

## CONCLUSIONS

The preparation of low temperature  $\text{TiO}_2$  photoelectrodes have been applied in fabrication of flexible dye-sensitized solar cells. The best performance of Dye Sensitized Solar Cells (DSSC) resulted on the thermal annealing of  $120 \text{ }^\circ\text{C}$  for 4h. The conversion efficiency of 0.019 % was obtained with a short circuit photocurrent of  $3.94 \times 10^{-2} \text{ mA}$ , an open circuit voltage of 0.938 Volt, and a fill factor of 0.53 under illumination

with 50 mW/cm<sup>2</sup>. The improvement of cell performance by applying some post treatments, controlling the thickness, static pressing, and microwave irradiation in the preparing of nanoporous TiO<sub>2</sub> particles can be expected.

## ACKNOWLEDGMENT

This work was supported by the State Ministry of Research and Technology under Sinas Programs 2012.

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