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by Jatmiko Endro Suseno

Facile method for synthesis of TiO_2 film and its application in high efficiency dye sensitized-solar cell (DSSC)

Abstract. Dye-sensitized solar cells (DSSC) is a device which converts a solar energy to electrical energy. Different with semiconductor thin film based solar cell, DSSC utilize the sensitized-dye to absorb the photon and semiconductor such as titanium dioxide (TiO_2) and zinc oxide (ZnO) as a working electrode photoanode. In this report, the preparation of TiO_2 film using a facile method of spray deposition and its application in DSSC have been presented. TiO_2 photoanode was synthesized by growing the droplet of titanium tetraisopropoxide diluted in acid solution on the substrate of conductive glass flourine-doped tin oxide (FTO) with variation of precursor volume. DSSC was assemblied by sandwiching both of photoanode electrode and platinum counter electrode subsequently filling the area between these electrodes with triodine/iodine electrolite solution as redox pairs. The characterization of the as prepared DSSC using solar simulator (AM 1.5G, 100 mW/cm^2) and I-V source meter Keithley 2400 showed that the performance of DSSC was affected by the precursor volume.. The overall conversion efficiency of DSSC using the optimum TiO_2 film was about 1.97% with the open circuit voltage (V_{oc}) of 0.73 V, short circuit current density (J_{sc}) of 4.61 mA and fill factor (FF) of 0.58.

Keywords: TiO_2, Spray Deposition, Solar cell, Dye-sensitized solar cell, Photoanode.
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INTRODUCTION

Dye sensitized solar cells (DSSCs) is a new generation of solar cells. Since it was discovered by Gratzel et al. [1], the research and development of DSSC increase rapidly. The DSSC is different from the commercially silicon-based solar cell since in the former cell there is a separation between light absorption function and carrier transport function [2]. DSCC-type solar cell has many advantages, that is including easy to be produced, cheaper cost of production than the silicon-based solar cell and has a high efficiency even though in the low intensity of light[3]. In general, DSSC comprise of photo-anode which is made from semiconductor such as TiO_2 and ZnO, dye as sensitizer which absorb sunlight, a pair of redox in the form of triodide/iodide, and electrode counter from platinum (Pt) [1].

A thick film of TiO_2 was produced by coating the nano-particle TiO_2 paste on top of a transparent conductive glass (TCO glass) using doctor blade and screen printing methods. These methods were proven to be able to produce TiO_2 nanoparticle film which has homogeneous thickness. However, it was required three steps to produce a TiO_2 nanoparticle film. The first step was producing TiO_2 nanoparticle. The second step was to produce TiO_2 paste and to disperse the obtained TiO_2 nanoparticle in the appropriate liquid medium, and then followed by the addition of the appropriate surfactant. The last step is coating and annealing.

In our work, the photo-anode electrode was produced using spray-deposition method which was also called aerosol based method. This simple method was capable of producing a film which has a controllable thickness with a high homogeneity [4-5]. A controlled film thickness allows repeating the production to obtain the films with the high consistency. A high efficiency of DSSC required the optimum thickness of film in photo-anode TiO_2, since it was affecting light absorption process [6]. In this paper, we study the optimum condition for TiO_2 film coating. We also observe the effect of that condition in the resulted photo-to-current conversion efficiency (PCE) and photovoltaic DSSC parameter.

METHOD

Production of TiO_2 photo-anode

TiO$_2$ photo-anode film is a thick film from TiO$_2$ which was coated on top of F-doped SnO$_2$ glass (FTO, TEC15, Dyesol). This film has sheet resistance ~15 ϕι/sq, transmittance ~80%, and glass thickness around 2.3 mm. The FTO coated glass was cleaned up using methanol, aceton and destilled water by ultrasonic process.. A TiO$_2$ film was prepared using spray deposition method as illustrated in Fig.1. The equipments consisted of dropet generator and temperature controllable hot plate. The precursor of prekursor TiO$_2$ was titanium tetraisopropoxide (TTIP, Aldrich, Singapore) which was dissolved in the liquid of HNO$_3$ (Merck, Singapore) 1 N. The concentration of TTIP liquid was preserved in 0.1 M. The next step was atomising the precursor liquid using droplet generator/sprayer. The resulted droplet was then transported on the surface of FTO glass which was heated at 500°C. The active area of TiO$_2$ layer was set at the length of 4 cm and width of 1 cm. In order to obtain a good quality of film with the appropriate thickness, the volume of precursor was varied. The quality of film will affect performance of the DSSC.

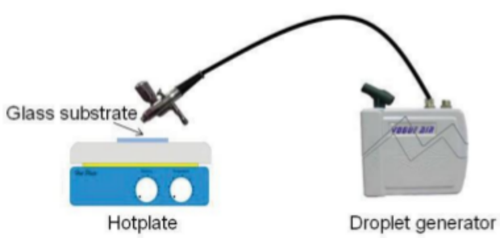

FIGURE 1. The scheme of spray deposition equipment.

The Assembly of DSSC

Components of DSSC consist of counter electrode from platinum (Pt) as illustrated in Fig.2a., TiO$_2$ photo-anode electrode (see Fig.2b.), sensitizer dye, a pair of redox electrolyte and plastic sealant. TiO$_2$ photo-anode electrode was submerged in the liquid of dye Ru (II)-complex (N719, Dyesol) 0.3 mM with dissolve acetonitrile:tert-butanol (1:1 vol:vol) for 24 hours. The counter electrode Pt was created by coating hexachloroplatinic acid based paste on top of FTO glass with the dimension which was the same as the dimension of the active TiO$_2$ layer using doctor blade method. After the paste was coated on top of FTO glass, then the electrode was annealed at 400 °C for 30 minutes. A dye coated TiO$_2$ film was piled up with the counter electrode where thermo-plastic was placed between them. Thermo-plastic has functions as spacer *sealant* (Surlyn-30, Dyesol). The slit between electrodes was filled with triiodide/iodide (I_3^-/I^-, Dyesol) as a pair of redox. The shape of DSSC is illustrated in Figure 2(c-d).

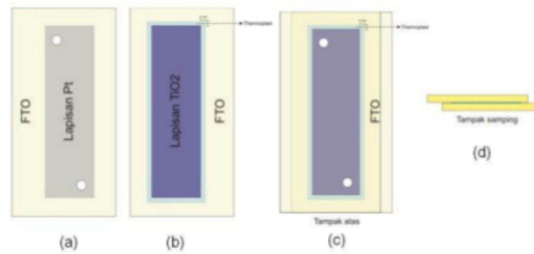

FIGURE 2. The scheme of (a) Pt counter electrode and (b) TiO$_2$ photo-anode. The view of DSSC scheme from (c) top and (d) side.

Characterization

Crystal structure of the obtained TiO$_2$ film was characterised using X-ray diffractometer (XRD, RINT 2200V, Rigaku-Denki Corp., Tokyo, Japan). The measurement was performed using nickel-filtered CuK$_\alpha$ radiation (λ= 0.154 nm) at the voltage of 40 kV and current of 30 mA with scan step of 0.02° and scan speed of 4°/min). A performance test of DSSC was carried out using solar simulator (AM 1.5G, 100 mW/cm2; L01, Peccell Technology, Japan) and source meter Keithley 2400. The intensity of incident solar illumination was set at the condition of 1 sun with the certified reference of Si cell attached to the KG-5 filter.

RESULT AND DISCUSSION

In this work, DSSC photo-anode in the form of TiO$_2$ film on top of FTO glass was grown with the precursor volume varied as 5, 7, 15 and 20 mL. The result of the crystal structure measurement using XRD for the 7 mL precursor is illustrated in Figure.3. The XRD analysis indicates that the produced TiO$_2$ has anatase phase with the diffraction peaks can be represented by the crystal orientation planes of (101), (004), (200), (211) and (118), which correspond well to the data in JCPDS file No. 84-1286. The other diffraction peaks belong to SnO$_2$, which is compatible to to the data in JCPSD file No. 77-0451. It is clear that by setting the annealing temperature at 500 °C the good crystallites of TiO$_2$ film is created which in turn it is expected that it can transport injected electron efficiently. Hence, it affects the performance of DSSC [7].

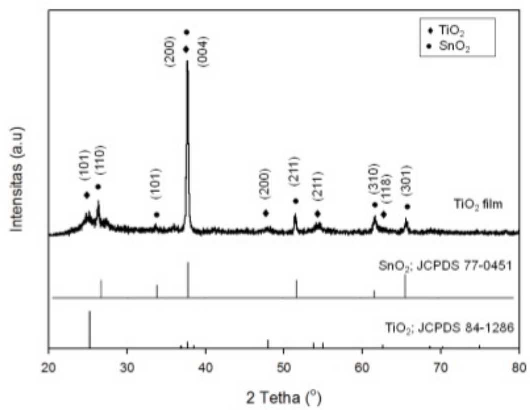

FIGURE 3. The XRD pattern of the TiO_2 which is grown on top of FTO glass.

Figure 4 shows the SEM images of the TiO_2 film. The active area of TiO_2 film was adjusted at 4 cm^2. From these images, it was clear that the film was formed on the surface of FTO glass. However, increasing the precursor volume from 5 to 15 mL tend to form a cracked film. The possible reason to explain this phenomena is increasing the volume may decreasethe adhesive force of TiO_2 on the SnO_2. Additionally, the heating and cooling process during the formation of TiO_2 was naturally carried out without adjusting the heating rate.

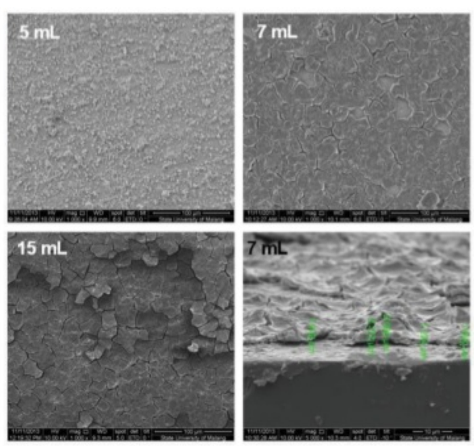

FIGURE 4. The SEM images of the TiO_2 which is grown on top of FTO glass at different precursor volume.

The TiO_2 photo diode which is already assembled with the other DSSC components is shown in Figure 5. The resulted DSSC has TiO_2 working electrode with the dimension of 4 cm x 1 cm, while the dimension of FTO glass is 5 cm x 2,5 cm. The result of the DSSC performance test of the of the DSSC, performed at solar illumination simulator AM 1,5G, which is equivalent with 100 mW/cm^2, is shown as characteristic curve of photo-current density versus photo-voltage (J–V) in Figure 6. The photo-voltaic parameters is summarised in detail inTable 1.

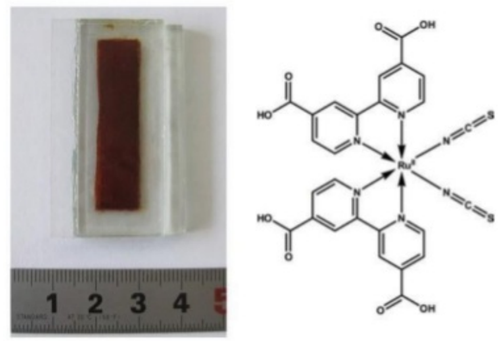

FIGURE 5. DSSC was created with Ru (II)-complex (N719) as its dye sensitizer.

The open-circuit voltage of DSSC using TiO_2 photo-anode with 5 mL precursor is 0.47 V and it is increase as the precursor volume increase.
It is seen that for TiO_2 with precursor volume of more than 7 mL, the V_{oc}- is around 0.7 V. However, the current density of the short-circuit (J_{sc}) is showing different trend, where in TiO_2 - DSSC with 7 mL volume precursor shows the maximum result as 4.61 mA/cm^2. Since the produced film using 7 mL precursor has optimum thickness, then it is expected that it will also havean optimum light absorption [6]. In general, the increase in precursor volume will directly increase the thickness of the film linearly. It is also affecting the quality of the film, for example due to the existence of micro-crack which decrease adhesion characteristic of the film on the FTO substrate [6]. Hence, it is clearly seen that by increasing the precursor volume above 7 mL, the current density J_{sc} will decrease. The values of fill factor (FF) in TiO_2 - DSSC with 5 mL and 20 mL precursor are relatively higher than the FF values of the DSSC with 7 mL and 15 mL precursor. This FF results are related to the ratio between maximum power and the product of open-circuit voltage and short circuit current density as in equation 1 below

$$FF = \frac{V_{\max} J_{\max}}{V_{oc} J_{sc}} \qquad (1)$$

It is clearly seen that the small V_{oc} and J_{sc} result in the high value of FF. This will affect photo to current conversion efficiency, PCE, ψ which is written as

$$\psi = \frac{P_{\max}}{P_{in}} 100\% = \frac{V_{oc} J_{sc} FF}{P_{in}} 100\% \qquad (2)$$

where P_{max} is maximum power, V_{max} and J_{max} represents voltage and current density at maximum power. In equation above, P_{in} is incident power which in this work is 100 mW/cm^2. In TiO_2 - DSSC with 7 mL precursor yield maximum PCE which is 1.97 %.

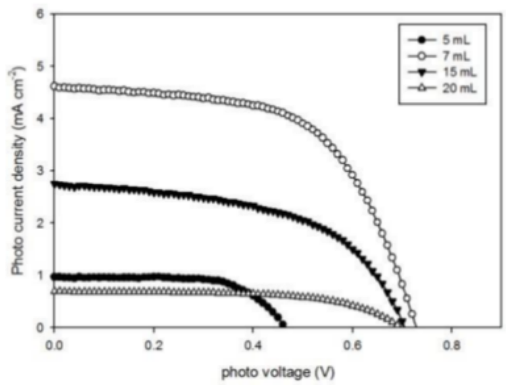

FIGURE 6. Characteristic curve J–V of DSSC with photo-anode TiO_2 where the precursor volume is varied.

TABLE 1. Photovoltaic parameters from the measurement of DSSC performance with illumination of AM 1,5G where the precursor volume is varied.

Precursor Vol. (mL)	V_{oc} (V)	J_{sc} (mA)	FF	ψ (%)
5	0.47	0.97	0.66	0.30
7	0.73	4.61	0.58	1.97
15	0.71	2.75	0.53	1.04
20	0.70	0.69	0.61	0.29

CONCLUSIONS

In this work, photo-anode TiO_2 has been successfully created by using spray-deposition method. The precursor volume influence TiO_2 film where the resulted film is a crystal with anatase phase. It also has an effect on resulted photo-to-current efficiency (PCE). DSSC with 7 mL precursor has the biggest efficiency which is 1.97 % with V_{oc} 0.73 V and J_{sc} 4.61 mA. These results show that appropriate precursor generates TiO_2 film with optimum thickness.

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