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Erlyta Septa Rosa
Research Center for Electronics and Telecommunication, Indonesian Institute of Sciences/LIPI, Bandung 40135, Indonesia, erlytasr@gmail.com

Lia Muliani
Research Center for Electronics and Telecommunication, Indonesian Institute of Sciences/LIPI, Bandung 40135, Indonesia

Shobih Shobih
Research Center for Electronics and Telecommunication, Indonesian Institute of Sciences/LIPI, Bandung 40135, Indonesia

Jojo Hidayat
Research Center for Electronics and Telecommunication, Indonesian Institute of Sciences/LIPI, Bandung 40135, Indonesia

Brian Yuliarto
School of Physics Engineering Faculty of Industrial Engineering, Institut Teknologi Bandung, Bandung 135 Indonesia

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Series-Interconnected Plastic Dye-Sensitized Solar Cells Prepared by Low-Temperature Binder-Free Titania Paste

Erlyta Septa Rosa[1*], Lia Muliani[1], Shobih[1], Jojo Hidayat[1], and Brian Yuliarto[2]

1. Research Center for Electronics and Telecommunication, Indonesian Institute of Sciences/LIPI, Bandung 40135, Indonesia
2. School of Physics Engineering Faculty of Industrial Engineering, Institut Teknologi Bandung, Bandung 40135 Indonesia

[*]*e-mail: erlytasr@gmail.com*

Abstract

The aim of this research is to study dye-sensitized solar cells (DSSC). This was implemented on a flexible polyethylene terephthalate (PET) substrate using a mixture of transparent and scattered mesoporous anatase-titania as the electron transport layer for the photoelectrode. This mixture of anatase titania performed a dual function of light scattering and efficient dye absorption. In this study, a porous nano-TiO_2 film was prepared on indium tin oxide (ITO) coated polyethylene terephthalate (PET) by using a binder-free titania paste; on it, a DSSC was fabricated. The paste which contained a mixture of TiO_2 nanoparticles, acid chloride, and ethanol was printed on two patterns of 1x6 cm^2 active areas followed by sintered at 120 ºC to form TiO_2 films. A commercial dye, N719, was adsorbed on the surface of TiO_2 films and assembled to two platinized conductive plastic patterns to form a counter electrode and thus a sandwich-type dye cell. Finally, a solution of KI/I_2 electrolytes was injected into the cell in which a couple of sandwich-type dye cells with an active area of 6 cm^2 for each cell were series interconnected with a z-type interconnection between the photoelectrode of one cell and the counter electrode of another cell. The cell performance was characterized by employing simulated solar light at an intensity of 50 mW/cm^2. The results showed interconnected cells generating a short-circuit photo-current density of 2.34 mA/cm^2, an open-circuit voltage of 1.10 volt, and overall 0.172% power conversion efficiency.

Abstrak

Sel Surya *Dye-Sensitized* Plastik Terinterkoneksi Seri Menggunakan Pasta Titania *Binder-free* pada Temperatur Rendah. Tujuan dari penelitian ini adalah untuk mempelajari teknologi pembuatan sel surya berbasis *dye-sensitized* (DSSC). DSSC dibuat pada substrat fleksibel polyethylene terephthalate (PET) menggunakan campuran mesopori anatase-titania transparan dan *scattered* sebagai lapisan transpor elektron (fotoelektroda). Campuran anatase titania tersebut mempunyai fungsi ganda yaitu sebagai penghambur cahaya sekaligus sebagai penyerapan zat warna yang efisien. Dalam studi ini, fotoelektroda DSSC dibuat menggunakan film TiO_2 nano berpori yang dilapiskan di atas substrat PET yang berlapis indium tin oxide (ITO) menggunakan pasta titania binder-free. Pasta yang terdiri dari campuran TiO_2 nanopartikel, asam klorida, dan etanol dilapiskan pada dua pola dengan daerah aktif masing-masing 1x6 cm^2 dilanjutkan dengan proses sintering pada temperatur 120 ºC. Subtrat direndam dalam larutan pewarna komersial, N719, sehingga pewarna teradsorpsi pada permukaan film TiO_2. Kemudian substrat digabungkan (*assembling*) dengan substrat PET yang telah dilapisi platina sebagai elektroda *counter* sehingga membentuk *sandwich*. Akhirnya, larutan KI/I_2 elektrolit disuntikkan di antara celah kedua elektroda di mana dua buah sel surya dengan area aktif masing-masing 1x6 cm^2 terinterkoneksi secara seri dengan pola z untuk menghubungkan fotoelektoda salah satu sel dengan elektroda *counter* sel lainnya. Pengukuran kinerja sel dilakukan dengan menyinari sel menggunakan simulator matahari pada intensitas cahaya 50 mW/cm^2. Hasil penelitian menunjukkan sel-sel yang terinterkoneksi tersebut menghasilkan arus hubung singkat 2,34 mA/cm^2, tegangan rangkaian terbuka 1,10 volt, dan efisiensi konversi daya sebesar 0,172%.

Keywords: DSSC, photoelectrode, series interconnected, TiO_2 film, titania paste

1. Introduction

The dye-sensitized solar cell (DSSC) is a type of photo-electrochemical solar cell composed of a sensitizing dye, a nanoporous wide-band semiconductor photo electrode, a counter electrode, and an electrolyte containing a redox couple [1,2]. When a DSSC is illuminated by sunlight, the dye molecules, which are anchored to the

surface of the wide-band semiconductor, absorb light and become excited. The absorption of light by the dye molecules is followed by the injection of an electron from the excited state of the dye to the conduction band of the semiconductor and its subsequent transfer to the transparent oxide. Finally, the electron flows through the external circuit [1]. In the DSSC, the photo electrode plays an essential role, and usually the photo electrode is a TiO_2 nanocrystalline film comprising a three-dimensional network of interconnected 15–20 nm-sized nanoparticles sintered onto a conductive glass [2]. Currently, many laboratories are undertaking research on DSSCs due to their relatively high efficiency and potentially low-cost production [3]. The best efficiencies have been measured on a very low geometrical area (about 0.25 cm^2) [2].

Transparent conducting oxide (TCO) coated glass is usually employed as a substrate material for DSSC due to its excellent optical, electrical, and encapsulation barrier properties, as well as its compatibility with high-temperature processing [4]. Plastic DSSCs based on the substrates of indium tin oxide (ITO) coated polyethylene terephthalate (PET), or polyethylene naphthalate (PEN) substituting for a rigid glass substrate, are regarded as possible breakthroughs in the field of DSSC in terms of commercialization because plastic DSSCs have the advantages of low cost of production and wide application [5]. Large-scale roll-to-roll printing process can be applied to the fabrication of plastic-substrate DSSC, making its commercialization more viable [6].

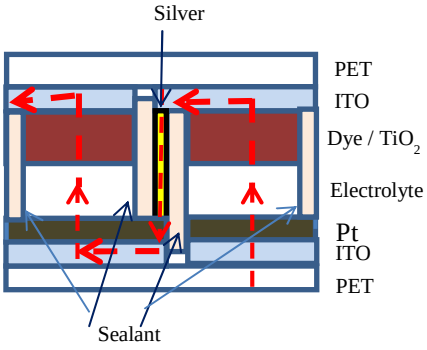
Underlying the plastic DSSCs, the necessary low-temperature preparation of porous nanocrystalline metal oxide semiconductor films has been an ongoing challenge because the conventional method of high-temperature preparation cannot be applied to films on plastic substrates, which can only endure a temperature of up to around 150 ºC. So far, there have been a number of efforts concerned with the preparation of nanoporous TiO_2 films at low temperatures [5]. The methods reported were low-temperature heating, compression, microwave irradiation, electron-beam annealing, chemical-vapor deposition with UV irradiation, and hydrothermal crystallization [5]. Electrophoretic deposition of TiO_2 followed by chemical necking treatments and vacuum cold spraying at room temperature were introduced by Kijitori and Fan [4,7]. However, the conversion efficiencies of the flexible DSSC achieved so far are lower than those obtained by high-temperature sintering.

Conventionally, TiO_2 film is prepared by coating a titania paste containing binder and organic additives, followed by a procedure of high-temperature sintering to remove the binder and organic additives. The high-temperature sintering is also necessary to form a thick crack-free uniform film and optimize the microstructure of the photo electrode for photosensitization [5]. Meanwhile, this high-temperature processing is only suitable if using ITO coated glass as a substrate.

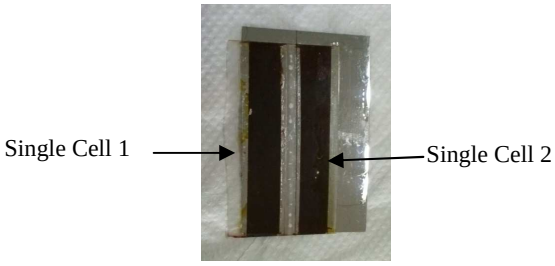
The aim of this research is to study DSSC implemented on a flexible PET substrate using a mixture of transparent and scattered mesoporous anatase titania as the electron transport layer for the photoelectrode. This mixture of anatase titania performed a dual function of light scattering and efficient dye absorption. In this study, a porous nano-TiO_2 film was prepared on an ITO coated PET, by using a binder-free titania paste, on which a DSSC was fabricated. The paste which contained a mixture of TiO_2 nanoparticles, acid chloride, and ethanol was screen printed on two patterns with 1x6 cm^2 active areas, followed by sintered at 120 ºC in a vacuum oven to form TiO_2 films. A couple of sandwich-type dye cells with an active area of 6 cm^2 for each cell were series interconnected with a z-type interconnection between the working electrode and the counter electrode. The cell performance was characterized by employing simulated solar light at an intensity of 50 mW/cm^2. It is shown that TiO_2 films from a binder-free titania paste can work effectively in plastic DSSC.

2. Experiment

A binder-free titania paste was prepared using the following method: 6 gr nc-TiO_2 (91% transparent-anatase, particle size 20 nm, and 9% scattered-anatase, particle size 150–250 nm) from Dyesol, 3 ml ethanol (Merck), a few drops of 1 M HCl water solution, and Triton X-100 as a surfactant were ground in an agate mortar for about 2 h to get a viscous paste. A 9% scattered-anatase with 150–250 nm particle size was used to lengthen the optical path length in the TiO_2 film [8]). ITO coated PET substrate (sheet resistance 60 ohm/sq) with a dimension of 4 x 7 cm^2 was grooved laterally near the middle of the substrate to isolate two electrodes for respective cells. The substrate was then cleaned by soaking it in a detergent solution, H_2O, and IPA, respectively, for 10 minutes in an ultrasonic condition. After drying the substrate, the titania paste was then coated on two patterns of 1x6 cm^2 ITO coated PET (sheet resistance 60 ohm/sq) using a doctor-blade technique. Subsequently, the film was sintered at 120 ºC for 4 h to form TiO_2 photo electrode. The TiO_2 layer was directly immersed in an ethanol solution of ruthenium (II) based dye N719 (Dyesol) overnight at room temperature. This dye-sensitized electrode was employed as the working electrode. For the counter electrodes, the cleaned ITO PET substrate with dimensions of 4x7 cm^2 was coated by a thin film of platinum by sputter deposition in a vacuum. The substrate was then grooved laterally near the middle of substrate to isolate two patterns of counter electrodes by removing ITO and platinum layers. Series-interconnections of two plastic DSSC were formed by arranging



(a)



(b)

Figure 1. The Structure of a Series Interconnected with a Z-type Interconnection between the Working Electrode and the Counter Electrode (a) and the Picture of the Cell (b)

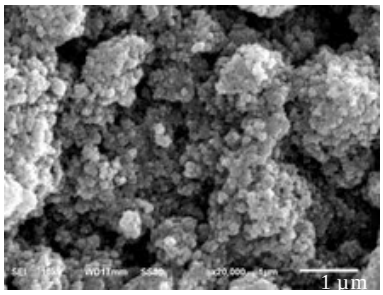
working electrodes and counter electrodes in a sandwich and placing spacers (surlyn thermoplastic sealant, Dyesol) and silver paste to form a z-type interconnection between working electrode of one cell and counter electrode of another cell (see Figure 1). The cells were then heated at 120 °C for 10 minutes to cure the paste and spacers. The solution of KI/I_2 electrolyte in 3-methoxypropionitrile (Dyesol) was injected into the cells through two holes (one at the top and one at the bottom) between the spacers in each cell. The holes were then sealed with surlyn thermoplastic sealant (Dyesol). For electrical measurement, the cells were tested using an I-V measurement system under a sun simulator from a xenon light equipped with an AM 1.5 filter at an intensity of 50 mW/cm^2. The morphology of the TiO_2 photoelectrode was observed using a scanning electron microscope.

3. Results and Discussion

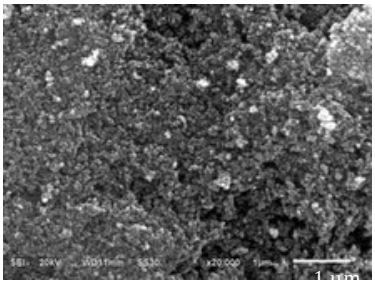
TiO_2 working electrodes used in DSSC are composed of 20 nm size crystallites. These electrodes are essentially transparent since visible light is not scattered for titania particles of sizes in the order of a few tens of nanometers. In fact, incident photons that are not absorbed by the dye-sensitized electrode are either lost through the counter electrode or partially absorbed by the electrolyte solution. The first attempts to collect these escaping photons were based on the use of polydisperse packings of sub-micron size spheres as highly diffusive reflecting layers [8].

The scanning electron microscopy (SEM) image of the TiO_2 photo electrode after sintering at a low temperature of 120 ºC is presented in Fig. 2. The SEM images of the transparent-anatase and scattered-anatase TiO_2 are also presented in this figure for comparison.

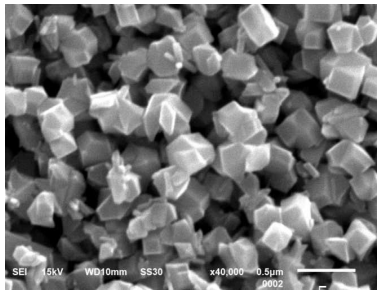
As shown in Fig. 2(a), the film is composed of random-size agglomerates. The agglomerates are arranged from large particles of scattered-anatase TiO_2 and small particles of transparent-anatase TiO_2. The agglomerate consist of a mixture of 91% transparent-anatase TiO_2 (particle size 20 nm) and 9% scattered-anatase TiO_2 (particle size 150–250 nm). The aggregates can be designed to generate effective light scattering and thus extend the traveling distance of light within the photoelectrode film [9].



(a)



(b)



(c)

Figure 2. SEM Images of (a) TiO_2 Photoelectrode After Sintering at 120 ºC, (b) Transparent-anatase nc-TiO_2, and (c) Scattered-anatase nc-TiO_2

The uneven distribution of scattered-anatase and transparent-anatase TiO_2 in the agglomerates indicates that the titania paste was not spread homogenously due to the short time available for mixing. The image shows that there are many formations of porous surfaces on the TiO_2 film, indicating the film has a high surface area. The high surface area of TiO_2 film is needed for the adsorption of a large number of dye molecules and for an electrical connection with the redox electrolyte [10]. However, the interconnection between particles and the chemical bonding at the interface of the substrate may be poor because of the random agglomerates and the low-temperature sintering. It can be observed that the film was immersed in dye solution, as precipitation appears in the solution after the film is removed. As a consequence, the thickness of the TiO_2 film decreased, and the adsorption of the dye molecules was reduced, so that the absorption of the light is low.

The sandwich-type dye cell fabricated in this research is a combination of two single cells with an active area of 1x6 cm^2, which are series interconnected with a z-type interconnection between the working electrode of one cell and counter electrode of another cell. The photo current-voltage characteristic is shown in Figure 3. The detailed electrical characteristic of the cells are calculated from the figure and presented in Table 1.

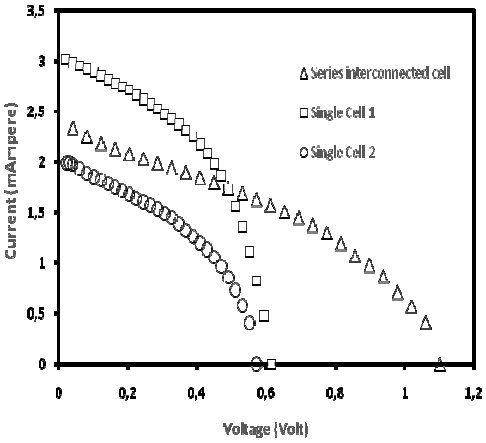


Figure 3. Photocurrent-voltage Characteristic of the Cells Under 50 mW/cm^2 Illumination

Table 1. Electrical Characteristic of the Cells Under 50 Mw/Cm^2 Illumination

Electrical characteristic	Series-interconnected	Single Cell 1	Single Cell 2
Voc (Volt)	1.10	0.61	0.57
Isc (mAmpere)	2.34	3.01	1.99
FF	0.39	0.48	0.43
PCE (%)	0.17	0.30	0.17
R_s (ohm)	223	54	105

The open-circuit voltage (V_{oc}) value of single cell 1 is quite similar to single cell 2. Unlike Voc, the short-circuit photo current density (I_{sc}) for single cell 1 is 1.5X higher than single cell 2. The difference in I_{sc} value may be due to the difference in the TiO_2 layer quality in each cell. In manual doctor-blade printing and inhomogenous pressure distributed on the blade may cause the TiO_2 layer density to be different in each cell but have the same thickness. In the case of single cell 1 and single cell 2 printing, if the blade pressure in on single cell 1 is higher in the single than cell 2, the TiO_2 layer on single cell 1 is also denser than on single cell 2 after sintering. Therefore, after sintering, the interparticles TiO_2 connectivity and the contact between the TiO_2 layer and substrate in single cell 1 are better than single cell 2. It means that electrons injected to the TiO_2 electrode in single cell 1 flow more easily than in single cell 2, which is indicated by their R_s values (54 ohm for single cell 1 compared to 105 ohm for single cell 2).

The I_{sc} for single cell 1 is 3.01 mA or 0.502 mA/cm^2, while the theoretical limited value of I_{sc} was reported at about 25 mA/cm^2 when all ranges of visible light (λ = 400–800 nm) under 1 sun irradiance were converted to external current in a single-cell [8]. The low I_{sc} for both cells is influenced by the higher charge recombination in the TiO_2/electrolyte interface where the electrons cannot collect and transport effectively. The high series resistance (R_s) also contributed to the low I_{sc}. High series resistance may be due to the weak electro-chemical bonding between the TiO_2 film and the substrate and the diffusion limit of electrolyte to the film interface. Another factor, which may contribute to the R_s is the high sheet resistance of the anode or ITO layer (60 ohm/sq.). As a result, the power maximum density (P_M) and the efficiency are lower than single cell 1. The low in P_m is shown in Figure 3, where the I-V curve area for single cell 2 is smaller than single cell 1. The efficiency was calculated as [2]

$$\eta(\%) = \frac{P_M}{P_S} = \frac{V_{oc}\, j_{sc} FF}{P_S} \quad (1)$$

where P_s stands for the power density of the incident illumination, and the fill factor is calculated by

$$\eta(\%) = \frac{2_d}{3}\sqrt{\frac{2kd}{3sA}} = 0.544d\sqrt{\frac{k}{C_0}} \quad (2)$$

where V_{mm} and J_{mm} are current density and voltage for maximum power output, respectively.

Baglio et al. [2] stated that the conversion efficiency appears to be related to reaction kinetic (R_{ct}). The reaction rate on the photo anode is strictly related to the number of oxidized dye species that are reduced by I^-

ions at the interface. This trend may also indicate that besides the recombination effect and absorption characteristics, an important role is played by the amount of light that effectively reaches the electrode-electrolyte interface to promote the dye transition from the ground to oxidized state [2].

Under 50 mW/cm^2 illumination, a series-interconnected cell generates an I_{sc} of 2.34 mA or 0.195 mA/cm^2, a V_{oc} of 1.10 V, and an FF of 0.39, yielding an overall 0.172% PCE. These values are a combination of the electric characteristics of single cell 1 and single cell 2.

The V_{oc} of series-interconnected cell is 1.10 V, which is an addition of each cell's voltage (0.61 V + 0.57 V). The I_{sc} of the series-interconnected cell is lower than single cell 1, but higher than single cell 2. However, the R_s is higher than that of single cell 1 and single cell 2. This may be due to the parasitic losses from contact between silver and ITO in both single cell 1 and single cell 2, so that the electron cannot flow effectively from cell 1 to cell 2.

The power conversion efficiency obtained is similar to the single cell 2, but is still lower than single cell 1. It seems that the efficiency of the series interconnected cell depends on the lowest efficiency of the single cell. The results reported by other researchers on flexible substrates show higher efficiency (1.7–2.5 %) compared with the results of this study (0.172%). However, they used very small active areas (order of mm) in single cell design [11,12].

This research is as a preliminary study in the fabrication of lightweight and flexible DSSC for mobile phone battery charging applications. A wider active area (2x6 cm^2 in this research) is needed to obtain suitable performance for mobile charging.

4. Conclusions

Porous TiO_2 films were prepared on an ITO coated PET using a binder-free titania paste at low-temperature sintering. The TiO_2 films were used as a photoelectrode for two DSSCs with an active area of 1x6 cm^2, which was series interconnected with z-type interconnection between the photoelectrode and the counter electrode.

The results showed that the series interconnected cells generated a short-circuit photo-current density of 2.34 mA or 0.195 mA/cm^2, an open-circuit voltage of 1.10 V, and overall 0.172% power conversion efficiency.

Acknowledgement

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