



# Article Influence of Coating a TiO<sub>2</sub> Electrode with DN-F05 and DN-F05-Ag on the Photovoltaic Performance of DSSC Solar Cells

Agus Supriyanto <sup>1,\*</sup>, Diani Galih Saputri <sup>2,\*</sup>, Mohd Khairul Bin Ahmad <sup>2</sup>, Artono Dwijo Sutomo <sup>1</sup> and Ari Handono Ramelan <sup>1</sup>

- <sup>1</sup> Department of Physics, Faculty of MIPA, Universitas Sebelas Maret, Surakarta 57126, Indonesia; artono@staff.uns.ac.id (A.D.S.); aramelan@mipa.uns.ac.id (A.H.R.)
- <sup>2</sup> Microelectronic and Nanotechnology-Shamsuddin Research Centre (MINT-SRC), Faculty of Electrical and Electronic Engineering (FKEE), Universiti Tun Hussein Onn Malaysia, Parit Raja 86400, Malaysia; akhairul@uthm.edu.my
- \* Correspondence: agusf22@staff.uns.ac.id (A.S.); dianigalih1@gmail.com (D.G.S.)

**Abstract:** A DSSC's performance depends on its working electrode. Since this working electrode functions as an electron carrier, it affects how well DSSC solar cells perform. This study focused on working electrodes that use the dye DN-F05 and the Ag ions in the hybrid dye DN-F05 as a sensitizer. This research was conducted using the spin-coating method. Based on structural, optical, and electrical data, the results obtained were related to each other. It was determined that Ag has a significant influence on DSSC performance, and this discovery was also the main goal of this study. Previous studies have shown that the addition of Ag as a doping agent is an effective strategy for improving chemical and electronic properties. This can be proven through electrical measurements, which have shown an efficiency of 5.33%. However, when a significant amount of Ag is hybridized in the dye DN-F05, it will cause a decrease in DSSC performance due to the accumulation of Ag particles on the seniconductor's surface, creating a barrier to light absorption and aggregation. This is what led to the selection of a hybrid material (Ag metal), as Ag is unique and offers great potential with respect to boosting solar cells' performance.

Keywords: dye DN-F05; Ag metal; TiO2 (anatase); spin coating

# 1. Introduction

In 1991, Gratzel and coworkers first proposed the idea of DSSCs [1]. Due to their ease of production, compatibility with architecture and the environment, and strong performance in diffuse light environments, dye-sensitized solar cells (DSSCs) have received a great deal of attention [2,3]. According to Galindo et al. [4], the functional mechanism of DSSCs is remarkably similar to photosynthesis in plants. The process of conversion is more complicated, involving the absorption of photons from radiation and their subsequent conversion to electrical energy.

DSSCs have straightforward structural designs and are typically composed of typically inexpensive materials. The components of a typical DSSC (Figure 1) are a transparent conducting oxide (TCO) glass substrate or fluorine-doped tin oxide (FTO), a wide bandgap semiconductor, a dye sensitizer attached to the surface of a semiconductor, and an electrolyte (DMPI traditional electrolyte) acting as a redox mediator and containing an iodine/triiodide ( $I/I_3^-$ ) redox couple [5]. However, in this study, we are more focused on constructing the working electrode. The DSSC anode side (semiconductor layer) is a material that has porous absorption [6], making it sensitive to the absorbed dye spectrum and granting it luminescence emission properties at wide wavelengths [7]. The n-type semiconductor materials commonly used for DSSC layers are ZnO, SnO<sub>2</sub>, In<sub>2</sub>O<sub>3</sub>, Nb<sub>2</sub>O<sub>3</sub>,



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and TiO<sub>2</sub> [8,9]. One of the most popular and frequently used semiconductor materials is TiO<sub>2</sub> [10–13].

Figure 1. Schematic illustration of a typical dye-sensitized solar cell (DSSC).

 $TiO_2$  is a semiconductor material with high porosity and a wide band gap at 3.2 eV [11,14,15]. The wide band gap value of  $TiO_2$  [16] lead to the active area limitation of the photocatalysis process, which only occurs in the UV light range. In recent years, to widen the wavelength of the  $TiO_2$  band gap, scientists have chosen a sensitizer that has a visible light wavelength area of 400–600 nm [6], allowing it to be used as a photon absorber [17].

A previous study reported that organic dye-based sensitizers using triphenylamine (TPA), which is commonly called dye D35 (Figure 2a), with a cobalt electrolyte produced high efficiency that was equal to 6.5% [18–20]. Ellis et al. [21] modified the conjugated linker by using different substitution dithiophene units to extend the absorption area of the D35 dye; it was determined that a high absorption area can increase photon absorption and the value of efficiency [22]. Combination with suitable bifunctionalized linker fragments has led generate to the synthesis of new dyes, namely, LEG1 (bithiophene), LEG2 (5-(thiopen-2-yl)-2,3-dihydrothieno [3,4-b] [1,4] dioxine), LEG3 (3-hexyl-2,2'-bithiophene), and LEG4 [DN-F05] (Figure 2b) (4,4-dihexyl-4H-cyclopenta [2,1-b: 3,4-b'] dithiophene as the  $\pi$ -conjugated linker) [21,22]. The highest efficiency value was obtained with DN-F05, which was used as a sensitizer in this study.



**Figure 2.** (a) Molecular structure of Dyenamo orange (D35) and (b) Dyenamo red liquid (DN-F05), and molecular structure of DN-F05 dye.

In recent years, there have been many methods for improving the short-circuit current in cells [16] (performance of TiO<sub>2</sub>) besides the use of a sensitizer, such as via metal doping [9,23–25]. The doping on the working electrode of semiconductors with certain elements will optimize their electrical and optical properties. Silver (Ag) is an exquisite metal that can be catalytic, nontoxic, and cost-effective and that possesses good thermal and electrical conductivity. In addition, doping Ag is one of the methods for enhancing its chemical and electrical properties and is often used to enhance DSSC performance [26–29]. In this investigation, a  $TiO_2$ -film-making experiment was conducted in which  $TiO_2$  that was immersed into dye DN-F05 mixed with Ag. In this study, the spin-coating method was used at an annealing temperature of  $450 \,^{\circ}$ C. Dye DN-F05 immersion was expected to produce a film layer with a band gap energy value suitable for maximizing photocatalytic activity at the working electrodes. The crystal and molecular structures of the product resulting from immersion in Dye DN-F05 with the addition of Ag were obtained through X-ray diffraction and Raman spectroscopy characterization. Moreover, the morphological layers were observed using FESEM characterization, while the band gap energy was measured via UV–Vis spectroscopy using the Tauc plot method and an IV meter to determine efficiency.

# 2. Materials and Methods

## 2.1. Preparation and Manufacture of TiO<sub>2</sub> Paste

The preparations required prior to the construction of  $TiO_2$  paste and dye DN-F05 included cutting FTO (Fluorine Tin Oxide) glass into dimensions of 2 cm  $\times$  1.5 cm as needed. Then, the FTO glass was cleaned using an ultrasonic cleaner for 15 min and dried. The conductivity of the dried FTO glass was evaluated using a multimeter.

The TiO<sub>2</sub> paste used in this study was a white paste consisting of TiO<sub>2</sub> nanoparticles. Anatase was purchased from Dyenamo Company. The obtained layer was ~300 nm thick. Dissolution of TiO<sub>2</sub> paste was achieved by employing ethanol at a ratio of 1:1. The solution was heated on a hot plate, allowing the substances to mix homogeneously. After applying maximum heating, TiO<sub>2</sub> was placed onto the FTO substrate using the spin-coating method (1000 rpm/30 s). The TiO<sub>2</sub> paste was coated on the FTO glass and annealed at 450 °C.

# 2.2. The Immersion of Dye DN-F05 and Dye DN-F05-Ag

Organic dye (DN-F05) was purchased directly from Dyenamo Company (Sweden). Dye DN-F05 was synthesized from dye D35 (Figure 2a). Dye DN-F05 was selected because it has higher efficiency values at 5.8% than the LEG-Series (LEG1;LEG2;LEG3;LEG4 (DN-F05)), which has an active area of 0.5 cm  $\times$  0.5 cm [21,22]. There were two dye immersions created in this study, namely, pure DN-F05 and DN-F05-Ag. Pure DN-F05 was created by dissolving dye with ethanol, while DN-F05-Ag was synthesized by mixing Ag metal solution into DN-F05 solution (Figure 3). The mixed Ag concentration was  $4 \times 10^{-1}$  M, and it was made by dissolving 0.0172 g of Ag into 40 mL of ethanol.



Figure 3. The experimental scheme of TiO<sub>2</sub>:DN-F05 and TiO<sub>2</sub>:DN-F05:Ag photoanode preparation.

## 2.3. Characterization

2.3.1. Working Electrode Measurement Using X-ray Diffraction

X-ray diffraction (XRD) testing was carried out to identify and analyze the crystal structure of pure TiO<sub>2</sub>, TiO<sub>2</sub>: DN-F05, and TiO<sub>2</sub>: DN-F05-Ag. Then, the results of the test

were matched with the data from the Inorganic Crystal Structure Database (ICSD) and the Joint Committee on Powder Diffraction Standards (JCPDS). X-ray diffraction (XRD) was used to determine the crystal structure by determining the peaks of the sample and comparing them with standard peaks. The samples were characterized using PANalytical X "Pert" powder with Cu K $\alpha$  radiation ( $\lambda$  = 1.5418 Å) from the MINT-SRC laboratory. X-rays were produced through the emission of electron voltage. The pound targets Cu X-rays that interact with the material will experience optical phenomena such as diffraction scattering, reflection, or transmission.

## 2.3.2. Field Emission Scanning Electron Microscopy (FESEM)

The FTO-substrate-coated TiO<sub>2</sub>-coated FTO substrate, which had been immersed in dye (DN-F05 and DN-F05:Ag), was characterized using FE-SEM via a JEOL JSM-7600F to analyze particle size and surface morphology. Before testing, the sample was coated in gold and placed in a sample holder, so the resulting morphological image of the layer was enlarged to the nanometer range.

#### 2.3.3. Raman Spectroscopy

The vibration modes of samples were determined using Raman spectroscopy (Horiba Scientific Xplora Plus, Tokyo, Japan). In this test, a 532 nm laser with a measurement range of 0–1800 cm<sup>-1</sup> was used. In general, the principle behind Raman spectroscopy is the interaction between light and matter using monochromatic light in the form of a laser. In this study, a 532 nm laser was used. Raman scattering occurs when the laser passing through the sample shows a frequency shift, which can be lower or higher than the initial frequency. Consequently, the information that can be obtained consist of vibrations, rotations, or other low-frequency transitions in the molecules.

## 2.3.4. Energy Band Gap Measurement

Decreases in bandgap energy were calculated using a UV–Vis spectrometer (Shimadzu UV-1800, Kyoto, Japan) operating at a wavelength ranging from 200 to 800 nm. Samples were tested in the form of layers (pure TiO<sub>2</sub>, TiO<sub>2</sub>:DN-F05, and TiO<sub>2</sub>:DN-F05-Ag). The magnitude of the optical band gap energy was obtained from data on the optical transmittance pattern of energy by observing the absorption coefficient  $\alpha$  as a function of photon energy using the Tauc plot method.

#### 2.3.5. Photovoltaic Measurement

Measurements were made using an I-V meter (solar simulator 1.5 AM ORIEL Sol1A). This test was carried out using an illuminator lamp with a light intensity of  $1000 \text{ W/m}^2$ . Figure 4 shows the structure of the sandwich system on a DSSC exposed to light.



Figure 4. DSSCs component assembly.

The counter electrode used was Pt Paste from Dyenamo Company, while the electrolyte used was traditional electrolyte (DMPI).

# 3. Results and Discussion

# 3.1. X-ray Diffraction Analysis

In this study, the phase/structure of silver (Ag) was a face-centred cubic (fcc) structure, while the crystal structure of TiO<sub>2</sub> was tetragonal. Figure 5 shows XRD diffractograms of pure TiO<sub>2</sub>, TiO<sub>2</sub> soaked in dye DN-F05, and TiO<sub>2</sub> soaked in dye DN-F05 mixed with Ag solution. Pure TiO<sub>2</sub> (Figure 5) lattice planes were found to be consistent with JCPDS no. 21-1272. The pure TiO<sub>2</sub> semiconductor diffractogram shows the angles included in the anatase phase at 37.8°. There is only one clearly visible anatase peak, whereas the other two peaks are faintly visible and almost invisible because the TiO<sub>2</sub> layer on the glass substrate (~300 nm) was not as thick as the layer in the reference (JCPDS no. 21-1272). The appearance of two new peaks at angles of 68.9° and 70.3° indicates the presence of FTO glass substrates.



**Figure 5.** The result of X-ray diffraction (XRD) pattern analysis between pure TiO<sub>2</sub>, TiO<sub>2</sub>: DN-F05, and TiO<sub>2</sub>:DN-F05:Ag.

Furthermore, Figure 5 (TiO<sub>2</sub>:DN-F05:Ag) also shows the results of XRD characterization for the TiO<sub>2</sub> substrate immersed in dye DN-F05 mixed with Ag in large concentrations. Based on ICSD database No. 87-0720, the clear peak that appears at 37.8° is the peak of Ag. The appearance of these peaks indicates that the presence of Ag is stable on the crystal surface and does not disturb the crystal structure of TiO<sub>2</sub> [28] due to the high concentration of Ag that can replace the atoms in the TiO<sub>2</sub> molecule (Ag impurities enter in the TiO<sub>2</sub> crystal lattice both through the interstitial) and cause atomic defects, while some of the other Ag peaks decrease and become invisible. Moreover, the TiO<sub>2</sub>:DN-FO5:Ag pattern looks thicker than that of TiO<sub>2</sub>:DN-F05, indicating that some peaks of anatase TiO<sub>2</sub> and FTO have been removed. The possible reasons for this include the aggregation of the dye and Ag mixture. Several other Ag peaks decreased in intensity and are not visible; this is because excessively large Ag concentrations can replace atoms in TiO<sub>2</sub> molecules (Ag contamination in the TiO<sub>2</sub> crystal lattice through interstitials), resulting in atomic defects. This phenomenon is the main reason why the addition of excessively high Ag concentrations will reduce the performance of DSSCs. Additionally, the XRD test results for  $TiO_2$  soaked in DN-F05 dye (Figure 5.  $TiO_2$ : DN-F05) show the appearance of different peaks than those of the anatase structure at angles of 33.7° and 51.6°, which indicate the peaks of the FTO glass used. These results are in accordance with the ICSD standard for  $SnO_2$  No. 77-0452 (ICSD #039178). Dye DN-F05 is an amorphous organic dye synthesized from dye D35, thus rendering the peaks of dye DN-F05 undetectable during XRD testing. However, amorphous materials are absorbed more quickly. Therefore, the DN-F05 dye attached to the substrate will aid the maximum absorption of photons.

The full width at half-maximum (FWHM) value (Table 1), which will be used to determine the size of a crystal using the Debye Scherrer equation, was obtained from the XRD test data. Debye Scherrer equations were used to determine that the sample's average  $TiO_2$  crystal particle size was 37.178 nm, the dye-immersed  $TiO_2$  crystal size ( $TiO_2$ :DN-F05) was 22.142 nm, and the crystal size in the sample with the most Ag added was quite large at 52.667 nm. The large concentration of Ag damages the semiconductor layer, which can damage the crystal structure. This is because an excessively high Ag concentration will cover the top surface of the layer, which then tries to enter the semiconductor pores and replaces the position of the  $TiO_2$  molecule, resulting in atomic defects [29]. The smaller the crystal particle size, the easier it is to absorb on the surface. The data show that crystal size decreases when the  $TiO_2$  substrate is immersed in DN-F05 dye due to the high number of excited electrons and the decreasing crystal size of  $TiO_2$ :DN-F05. The performance of the DSSC was improved by the small particle size, which enabled absorption.

Sample	Pos. (2Th.)	Int. (%)	FWHM Radian (β)	βCcos θ	$\mathbf{D} = \mathbf{k}\lambda/\beta\mathbf{C}\mathbf{cos}\;\theta$	D (nm)
TiO <sub>2</sub>	25.1563 25.3778	100 96.21	0.004 0.004	$0.004 \\ 0.004$	37.144 37.211	37.178
TiO <sub>2</sub> :DN-F05	25.2763 33.7241	100 65.15	0.010 0.006	0.009 0.005	15.938 28.345	22.142
TiO <sub>2</sub> :DN-F05:Ag	33.770 51.509	100 91.710	0.007 0.003	0.006 0.02	24.271 81.062	52.667

Table 1. Crystallite size calculation table for TiO<sub>2</sub>, TiO<sub>2</sub>:DN-F05, and TiO<sub>2</sub>:DN-F05:Ag samples.

## 3.2. FESEM Analysis

The results of FESEM characterization for the pure  $TiO_2$  substrate and dye-soaked  $TiO_2$  (DN-F05 and DN-F05-Ag) can be seen in Figure 6a–c. Each image was enlarged  $10,000 \times$  to determine the changes in the layer structure. Figure 6a shows pure  $TiO_2$  morphology, where there are uneven spherical lumps, while Figure 6b shows the  $TiO_2$  substrate structure soaked in dye DN-F05. There are small balls attached to the FTO coated by  $TiO_2$ , which proves the existence of a  $TiO_2$  layer on the FTO substrate.



Figure 6. FESEM images of: (a) FTO:TiO<sub>2</sub>; (b) FTO:TiO<sub>2</sub>:DN-F05; (c) FTO:TiO<sub>2</sub>:DN-F05:Ag.

Figure 6c shows the characterization results for the  $TiO_2$  substrate that was immersed in dye DN-F05 mixed with the Ag solution. It can be seen that there has been a significant structural change due to the addition of Ag, which induces structural damage. This is highly related to the nature of the semiconductor material used, as soaking in a large concentration of Ag will cause changes in certain properties and thereby increase conductivity. This is because too many Ag particles will attempt to enter the semiconductor layer. If the concentration of Ag is high, this will lead to particle aggregation on the TiO<sub>2</sub> surface, which will clog the coating's pores and hinder the absorption process [29].

## 3.3. RAMAN Analysis

Characterization using Raman spectroscopy is used for the determination of vibrational modes. Based on Figure 7, for pure  $TiO_2$ , it can be seen that the peak spectrum of 148.06 cm<sup>-1</sup> is very clear. Additionally, there are peaks at 399, 520, and 639 cm<sup>-1</sup>. The data generated are in accordance with the results of previous studies [30], which showed one sharp peak and three other peaks that follow. The types of vibrational modes identified via Raman spectra are shown in Table 2.



Figure 7. Raman spectra of TiO<sub>2</sub>, TiO<sub>2</sub>:DN-F05, and TiO<sub>2</sub>:DN-F05:Ag.

Table 2. Raman TiO<sub>2</sub> vibration mode.

Wave Number (cm $^{-1}$ )	Vibration Mode
148	O-Ti-O symmetrical stretching vibration
405	Symmetrical buckling of O-Ti-O
520	Asymmetric buckling of O-Ti-O
642	O-Ti-O symmetrical stretching vibration

Source: Ramasany et al. [30].

The authors of [30] reported the vibrational modes of the wave numbers seen in Table 2. In Figure 7, we can see the structural similarity of the dye-soaked TiO<sub>2</sub> substrate (DN-F05 and DN-F05-Ag) with pure TiO<sub>2</sub>. There is a peak at 592.09 cm<sup>-1</sup> indicating COO deformation. This is possible because the concentration of dye DN-F05 is too large, which causes the dye's aggregation. Dye DN-F05 raises the peak to above 1000 cm<sup>-1</sup>. Unexpectedly, the Raman spectra of TiO<sub>2</sub>:DN-F05:Ag have no effect on the change in the shape of the graph. However, the most interesting phenomenon from the spectra is the decrease in the intensity of the peak at  $1000 \text{ cm}^{-1}$  following the immersion of TiO<sub>2</sub>:DN-F05-Ag. This proves the interaction between Ag and TiO<sub>2</sub> after immersion, which influences the effect of Raman resonance for TiO<sub>2</sub> [29]. In addition, the Ag weakens the peak, and the sample presents a local lattice defect [28]. Therefore, high addition of Ag will decrease efficiency because it blocks the absorption of photons.

## 3.4. Band Gap Energy (Optical Properties)

In all the characterizations above, there are optical data that strongly support the performance of the working electrodes tested. This is evidenced by the results concerning the energy band gap for the three surfaces (shown in Figure 8). Several previous studies [11,14,15] mentioned that the area of band gap energy for the TiO<sub>2</sub> layer is 3.2 eV, which, in the graph, shows the same value at 3.13 eV (Figure 8a). To enhance DSSC performance, a smaller band gap value is required in order to maximally absorb the photons, thus allowing for immersion in dye DN-F05 and Ag ion in the hybrid dye DN-F05. The results in Figure 8b show a decrease in the band gap energy value, indicating that the absorption of photons will be maximized, especially by hybridizing the Ag ion into the dye DN-F05.



Figure 8. Band gap energy surfaces of: (a) TiO<sub>2</sub>; (b) TiO<sub>2</sub>:DN-F05; TiO<sub>2</sub>:DN-F05:Ag.

## 3.5. Photovoltaic Performance

The photovoltaic performance of the sandwich-type cells using an organic dye (DN-F05) as a photosensitizer ([TiO<sub>2</sub>:DN-F05]:Ag/electrolyte [DMPII]/Pt counter electrode) was measured at 1 sun (100 mW cm<sup>-2</sup>) intensity. Figure 9 shows the J–V curves of the TiO<sub>2</sub> immersed in dye DN-F05 (TiO<sub>2</sub>:DN-F05) and TiO<sub>2</sub> immersed in the dye hybridized with Ag (TiO<sub>2</sub>:DN-F05:Ag) for 12 h.

Figure 9 shows that the addition of the dihybrid Ag into the DN-F05 dye increases electron transport, as indicated by the increasing value of Jsc, thereby helping to increase energy harvesting. This proves that the conductivity of  $TiO_2$  increased with the addition of Ag [16,29]. Therefore, the addition of Ag will increase the efficiency value.

Table 3 summarizes the photovoltaic parameters, such as Voc, Jsc, and FF, and the fabricated DSSC devices.



**Figure 9.** J-V characteristics of TiO<sub>2</sub>:DN-F05 and TiO<sub>2</sub>:DN-F05:Ag.

Table 3. Photovoltaic parameters of DSSCs.

Dye Concentrations	Voc (V)	Jsc (mA/cm <sup>2</sup> )	FF	η %
TiO <sub>2</sub> :DN-F05	0.746	10.422	0.48	3.79
TiO <sub>2</sub> :DN-F05:Ag	0.754	14.814	0.48	5.33

## 4. Conclusions

The crystal structure of a material with a large band gap energy can absorb photons; oppositely, if the band gap is too small, cell regeneration will occur more quickly. The performance of the improved DSSC employing synthetic dye (DN-F05) and Ag is the author's main concern in this study. From the data presented, it can be concluded that Ag plays a significant role in determining cell performance; a high concentration of Ag will lead to aggregation on the semiconductor's surface and complicate absorption, resulting in a decrease in DSSC performance. However, there is another factor that applies to cells with moderate Ag concentrations that can enhance performance. Ag particles will aid electron movement, allowing more electron hole pairs to be created and thus providing opportunities for energy harvesting. This is demonstrated by the optical and electrical data measurements, where the addition of Ag led to an efficiency of 5.33%.

**Author Contributions:** All authors contributed to the study's conception and design. Material preparation, data collection, and analysis were performed by D.G.S., A.S. and M.K.B.A. The first draft of the manuscript was written by D.G.S., and all authors commented on previous versions of the manuscript. A.D.S. and A.H.R. contributed to the analysis of the data and the check of the final draft. All authors have read and agreed to the published version of the manuscript.

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