Effect of N719 dye concentration on the Conversion Efficiency of Dye Sensitized Solar Cells (DSSCs)

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ABSTRACT

In this study, DSSCs with various concentrations (0.125, 0.25 and 0.5 mM) of N719 dye have been successfully prepared using simple steps. The X-ray diffraction results of the TiO₂ film showed that it is polycrystalline with anatase phase (tetragonal system) having crystallite size of about 12 nm. The absorbance spectrum of the TiO₂ film and N719 dye at various concentrations was recorded by Ultra Violet – Visible (UV-Vis) spectrophotometer. The energy band gap of the TiO₂ film calculated by Tauc’s formula was ~ 3.1 eV. The scanning electron microscope (SEM) image of the TiO₂ surface shows a spongy-like morphology having open pores which can enhance the adsorption of the dye molecules the transport of electrons. The atomic force microscope (AFM) image and the distribution chart of the grains of the TiO₂ film show that the grain size was~ 82 nm. The DSSC prepared using N719 dye concentration of 0.5 mM achieved the highest conversion efficiency (η) of 2.504 %, respectively. Subsequently, the enhancement in efficiency was ~ 86 % compared with the conversion efficiency of DSSCs prepared with at N719 dye concentration of 0.125 mM and 0.25 mM.

1. INTRODUCTION

The world increasing consuming of fossil fuels, resulting in pollution of the environment and global warming, has led to a more interest on renewable energy sources and their technology (Kaimo et al., 2013).

Dye sensitized solar cells (DSSCs) are photoelectrochemical, alternative energy source devices that convert light energy into electricity (Mohd et al., 2014). DSSCs were first proposed by O’Regan and Grätzel in 1991. DSSCs typically consist of nanocrystalline titanium dioxide photoanode film, dye molecules, redox electrolyte, and a counter electrode in a sandwich structure (Pirhadi et al., 2016) as shown in Figure 1. Solar radiation is absorbed by the dye resulting in the injection of holes and electrons inside the electrolyte and the TiO₂ layer, respectively. The injected electrons and holes move through TiO₂ and electrolyte to the electrodes and collected in the external circuit (Chang et al., 2015). DSSCs have attracted considerable attention in recent
years because of their low cost, simple fabrication, high incident solar light-to-electricity conversion efficiency, colorful natures, and potential economic advantages (Mohd et al., 2014; Brian and Michael, 1991).

One of the important parts in DSSCs is the dye. Ru complexes used in DSSCS have shown quality photovoltaic characteristics which led to higher than 10% solar cell laboratory conditions (Ahmad et al., 2013). The most used dye sensitizers with high photo and chemical stability is “cis-bis (isothiocyanato-bis (2,2’-bipyridyl-4,4’-dicarboxylato) ruthenium (II) bis-tetrabutylammonium” (N719). The N719 dye contains 2 carboxylic acids and 2 carboxylate groups (-COOH and COO-), the carboxylate functional groups act as joining agents to immobilize the dye on the nanocrystalline TiO\(_2\) surface. The adsorption of N719 by TiO\(_2\) layer alters the electronic communication between layer and the electrode, which is an important feature in DSSC (Jiajie et al., 2011).

![Figure 1: Schematic diagram of DSSC basic structure.](image)

The aim of this work is to prepare a dye sensitized solar cell by Doctor-Blade method and study the effect of dye concentration on the solar cell efficiency.

2. EXPERIMENTAL PROCEDURE

All the materials used in this work were supplied by Dyesol Company/Australia. The materials were as follows: Fluorine Tin Oxide (FTO) (sheet resistance 8 Ω/sq) coated glass substrates, TiO\(_2\) paste (18-NRT), N719 dye, organic solvent based electrolyte (EL-HSE), platinum paste, sealing material, Acetone, distilled water. For the preparation of the working electrode, FTO glass substrate with size of (2.5 cm × 2.5 cm) was used. The FTO glass was cleaned in ultrasonic bath for 5 minutes in distilled water and for 5 minutes in acetone. The TiO\(_2\) paste was deposited on FTO glass by Doctor-blade method and the thickness of the titania layer was determined by the thickness of scotch tape which has a thickness of 10 μm placed on the right and left sides of the conductive face of substrate. Then the scotch tape was removed and the films were left to dry for 30 minutes in a covered Petri dish. Thereafter, the films were annealed at 550 °C for 30 minutes in ambient atmosphere. Finally, the working electrodes were allowed to cool at room temperature. After cooling, the working electrodes were immersed in various concentrations 0.125 mM, 0.25 mM, and 0.5 mM of N719 dye solution for 24 hours. For the preparation of the counter electrode, two holes of 1 mm diameter were drilled to enable a later injection of electrolyte, and platinum (Pt) paste was deposited on conductive side of FTO glass by Doctor-blade method, and the electrodes were then annealed at 450 °C for 30 minutes in ambient atmosphere. This leads to homogenous distributed platinum with good catalytic activity. The working electrode and counter electrode were assembled into a sandwich structure using sealant gasket, with a thickness of 30 μm as spacer. The sealant gasket was placed around TiO\(_2\) paste and the counter electrode was put on it while the Pt film faces the TiO\(_2\). Finally, few drops of the electrolyte were injected through the holes in the counter electrode by a pipette, and the holes were sealed by plaster to prevent evaporation. The crystallite phase of TiO\(_2\) was identified by X-
ray diffractometer (Shimadzu 6000, Japan) using CuKα radiation (λ= 1.5406 Å). The surface morphology and topography of TiO₂ were investigated by SEM (JSM-7000F) and AFM (SPM-AA3000, Angstrom). The UV–Vis absorption spectra of the TiO₂ film and N719 dye were measured by UV–VIS–NIR spectrophotometer (Shimadzu, UV-1800). The photovoltaic performance of the DSSCs was measured using Keithley 2400 multimeter and tungsten halogen lamp. Based on I–V curve, the fill factor (FF) was calculated according to the formula:

$$\text{FF} (%) = \frac{P_{\text{max}}}{J_{\text{sc}} \cdot V_{\text{oc}}} \times 100$$  \hspace{1cm} (1)

Where $P_{\text{max}}$ is the maximum power density, $J_{\text{sc}}$ is the short circuit photocurrent density and $V_{\text{oc}}$ is the open-circuit photovoltage. The photoelectric conversion efficiency ($\eta$) was calculated according to the following equation:

$$\eta (\%) = \frac{J_{\text{sc}} \cdot V_{\text{oc}} \cdot \text{FF}}{P_{\text{inp}}} \times 100$$  \hspace{1cm} (2)

Where $P_{\text{inp}}$ is the incident power density.

3. RESULTS AND DISCUSSION

3.1. Structural Analysis

Crystalline characterizations of TiO₂ film prepared by Doctor-blade technique on glass substrate were carried out by X-ray diffraction (XRD). Figure 2 shows the XRD pattern of the TiO₂ film annealed at 550 °C. From the figure, it was confirmed that the TiO₂ layer material has anatase phase with polycrystalline structure according to the International Centre for Diffraction Data (ICDD) standard card no. (21-1272) (Vijayalakshmi and Rajendran, 2012). The diffraction peaks were indexed to the crystal planes (101), (004), (200), (105), (211), (204) and (215) and this result is in agreement with the results reported by Wang et al. (2015). The highest and strongest peak of TiO₂ thin film was at 2θ ≈ 25.4° corresponding to (101) direction. The crystallite size of TiO₂ film was calculated by Scherrer’s formula given by the following equation (Wei and Chang, 2011):

$$D = \frac{0.9 \lambda}{\beta \cos \theta}$$  \hspace{1cm} (3)

Where D is the crystallite size, λ is the X-ray wavelength of CuKα radiation, $\beta$ is the full width at half maximum and θ is the Bragg’s angle. The lattice parameters of the TiO₂ film are $a = 3.781$ Å and $c = 9.477$ Å, which are in agreement with the standard values (i.e., $a = 3.785$ Å and $c = 9.513$ Å) and the crystallite size is 12.4 nm.

3.2 Morphological analysis

The surface morphology of TiO₂ thin film was characterized by SEM. Figure 3 displays the SEM image of TiO₂ film of 10 µm thickness which has been deposited on the FTO glass after annealing at 550 °C for 30 minutes. The image of the TiO₂ surface shows a spongy-like morphology having open pores which can enhance the adsorption of the dye molecules the transport of electrons (Wu and Wang, 2012). The average particle size of TiO₂ NPs is about 20-40 nm. The small particles of
TiO$_2$ film have larger surface area and subsequently absorb more dye molecules and this may lead to improved DSSC performance.

Figure 3: SEM image of TiO$_2$ thin film at 30,000X.

3.3. (AFM) Results

The surface topography of TiO$_2$ film prepared by Doctor-blade method on FTO glass was studied by Atomic Force Microscope (AFM) technique. The 3-D AFM image and granularity cumulative distribution chart of TiO$_2$ film annealed at 550 ºC for 30 minutes are shown in Figures 4a and 4b respectively. The average grain diameter, average roughness and the root mean square roughness of the TiO$_2$ film were ~82.48 nm, 0.356 nm and 0.423 nm respectively.

3.4. Optical properties

Figure 5 demonstrates the UV–Vis absorption spectra of N719 dye solution with different concentrations in the wavelength range of (350-800) nm. From the figure, it can be observed that the increase in the dye concentration leads to increase in the absorbance. Moreover, the UV-Vis absorption spectra show that N719 dye has two absorption peaks at ~380 and 520 nm.

Figure 4 (a) 3-D AFM image of TiO$_2$ thin film, (b) Granularity Cumulative Distribution chart of TiO$_2$ thin film.
Figure 6 illustrates the UV–Vis absorption spectrum of TiO₂ film annealed at 550 ºC. From the figure, it can be noticed that the film has clear and sharp absorption edge at wavelength of ~ 350 nm.

The direct band gap of the TiO₂ thin film was determined by plotting \((\alpha h\nu)^2\) vs. \(h\nu\). The optical band gap \(E_g\) value is estimated by extrapolation of the straight-line portion of the plot to zero absorption edge as shown in Figure 7. From the figure, it was observed that direct optical band gap for annealed TiO₂ thin film was 3.12 eV.

Figure 7: Tauc’s plot of TiO₂ film.

3.5. Current voltage characteristics

Figure 8 shows the J-V characteristics of DSSCs prepared by TiO₂ film thickness of 10 μm annealed at 550 ºC for 30 minutes and various concentrations of N719 dye (0.125 mM, 0.25 mM and 0.5 mM). From the J-V curves, it was noted that “short–circuit current density” increases as the dye concentration increases. This result agrees well with the UV-Vis results (fig.5). This result suggests that as the dye concentration increases, the TiO₂ layer adsorbs enough dye and achieve higher efficiency. The DSSC with 0.5 mM dye concentration has high \(V_{OC}\) and \(J_{SC}\), subsequently obtained highest efficiency. However, at low dye concentration (0.125 mM), the adsorption is low and the \(V_{OC}\) and \(J_{SC}\) efficiency becomes lower. It can be also observed that \(V_{OC}\) changes with concentration variation which is thought to be due to reactivity and instability of N719 (Mohd et al., 2014).

Table 1 shows the photovoltaic parameters of the DSSCs prepared using different concentrations of N719 dye solution. The
maximum efficiency (η) of 2.504 % was achieved at dye concentration of 0.5 mM and the maximum efficiency enhancement was about 86%.

Figure 8: J-V characteristics of DSSCs prepared by various concentrations of N719 dye.

| N719 dye concentration (mM) | V_{OC} (V) | J_{SC} (mA/cm^2) | V_{max} (V) | J_{max} (mA/cm^2) | FF | η (%) | Efficiency enhancement (%) |
|----------------------------|------------|-----------------|-------------|-----------------|----|--------|---------------------------|
| 0.125                      | 0.36       | 1.289           | 0.212       | 0.545           | 0.249        | 0.231              |
| 0.25                       | 0.535      | 3.429           | 0.323       | 2.079           | 0.366        | 1.343              |
| 0.5                        | 0.585      | 5.197           | 0.420       | 2.981           | 0.412        | 2.504              | 86                |

4. CONCLUSIONS

In this study, DSSCs with various concentrations (0.125, 0.25 and 0.5 mM) of N719 dye have been successfully prepared using simple steps. The absorbance in the wavelength range of (350-800) nm shows that N719 dye with 0.5 mM concentration has highest absorption. The DSSC prepared with 0.5 mM demonstrated highest efficiency of 2.504 %. Subsequently, the enhancement in the efficiency was about 86%.

Conflict of Interest

The authors have no conflict of interest to declare.

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