Performance of dye sensitized solar cells fabricated using N-719 dye incorporated with ZnO layer

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Abstract

The development of a new type of solar cells including dye sensitized solar cells (DSSCs), where the flexibility and low weight of the DSSCs are the key properties, is in rapid progress and the efficiency of the devices has continuously improved in the last decade. In this work, DSSCs are fabricated using the N-719 industry standard dye and ZnO as the semiconductor layer. Various fabrication conditions including impregnation time, coating temperature and number of ZnO layers, have been investigated. It was discovered that higher impregnation time and temperature negatively affect the generated current but not the voltage. This is contradicted by the effect of the number of ZnO layers where both the generated current and voltage are affected. The efficiency of the device drops when the number of ZnO layers, the impregnation time and the temperature increases. The results have shown that the best performance of the DSSCs is obtained when a single layer of ZnO is used with the impregnation time and temperature of 2hrs and 25°C, respectively. The maximum energy conversion efficiency of 2.20% and the fill factor of 0.55 were calculated from this device. This work will, in the future, support the production of environmentally friendly solar cells that can be suitably integrated with the building in order to maximize solar energy utilization.

Keywords: *DSSCs, N-719, ZnO, efficiency*

1. Introduction

Solar energy has been classified to be one of the most reliable sources of renewable energy due to high energy potential, environmentally friendly, clean, cheap and ease of accessible. Solar energy can be used in two different forms, electrical and thermal energy. The conversion of sunlight to electrical energy requires solar cells [1]. Si-based solar cells, both crystalline and amorphous, are wildly accepted to be used as a device to harvest sunlight and generate electricity [2]. However, Si solar cells still provide several disadvantages such as high weight, low flexibility, ease of breaking as well as the complexity of the production of highly purified Si wafers [3]. These obstacles make the Si-based solar cells unsuitable for the future practical application where low weight and high flexibility solar panels are essential for building integration. There is a need for a new technology to produce new types of solar cells that can be used to replace the Si solar cell.

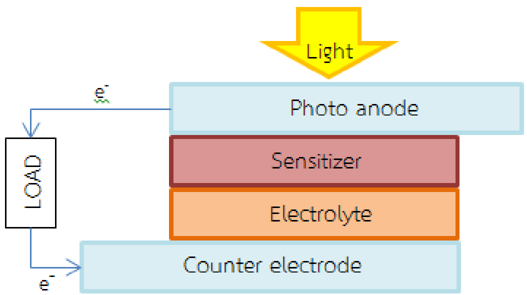

Figure 1 The fundamental structure of the DSSCs.

Dye sensitized solar cells (DSSCs) are expected to replaced Si solar cells in the near future [4] due to their suitability with integrating the solar panel and the building on both flat and curved surfaces. There has been vast interest in DSSCs fabrication technology due to their ease of production, and the potential to produce highly flexible low cost panels. This type of solar cell relies on the absorption of sunlight and the creation of charge carrier using the dye sensitizer molecules. The typical structure of a DSSCs is shown in Figure 1 where the device consists of a front electrode, photoanode, sensitizer active layer, electrolyte and counter electrode [5]. One main obstacle that prevents DSSCs from being realized in practical applications is their low efficiency. It has been shown that DSSCs fabricated using natural dye solutions provide the efficiency of about 0.2% [6] while a higher efficiency of 9.2% can be achieved using a synthetic dye sensitizer [7]. Another major problem of DSSCs is that the device shows low stability and can be seriously degraded after a long period of operation. This is due to the self-cleaning ability of the semiconductor particles when they interact with the ultraviolet radiation, leading to the degradation of the organic matter inside the cell [8].

The photoanode is made off a transparent conductive glass coated with a thin film of semiconductor nanoparticles. The structure of the semiconductor plays the key role in the electrical conversion process. Different types of nano-structured semiconductors are employed in the DSSCs fabrication including nano-rods, nano-tubes, nano-wires, nano-coneshelix and nanofiber [9] as a binding material between the transparent conductive glass and the dye molecules. Generally, TiO_2 is the primary choice of semiconductor nanoparticles selected to create the porous layer for dye molecules on the photoanode. However, ZnO is also another possible candidate to be used to form a semiconductor layer on the photoanode. A lot of researchers have turned their attention to ZnO because it offers several advantages over TiO_2. Even though the energy band gap of ZnO is very similar to that of TiO_2, it provides better charge transport compared to TiO_2. The preparation of a ZnO nanostructure can easily be accomplished and various nanostructures of ZnO can be selectively fabricated according to the requirement of the application. The charge transport ability of the DSSCs depends strongly on the characteristic of the semiconductor layer. Consequently, different structures of ZnO directly manipulate the efficiency of the DSSCs [10]. Therefore, ZnO is another viable choice of semiconductor layer to be applied as a free charge carrier generation and charge transport layer in the DSSCs. For the sensitizer, various types of dye molecules are used to coat the porous semiconductor layer on the photoanode side including N-719, N-3 and C-101. It has been shown that N719/ZnO device with different ZnO structure provides a different efficiency. ZnO nanowire structure exhibits the best performance with the conversion efficiency of 0.32% [11]. Size of ZnO particles is also important as it controls the conversion efficiency of the device. It was observed that the conversion efficiency of 4.54% can be obtained when the size of ZnO particle is in the range of 25-30 nm [12]. In this work, we explore the feasibility of using ZnO with N-719 dye sensitizer to construct the photoanode. The conditions for DSSCs fabrication are investigated including soaking time, soaking temperature and the number of ZnO layers in order to obtain the maximum efficiency of ZnO/N-719 DSSCs.

2. Experimental details

DSSCs are fabricated using the following materials. The transparent conductive oxide (TCO) glass, 3 mm thick, coated with fluorine-doped tin oxide (FTO) with a sheet resistance of 13 Ω/cm^2 was used to construct the photoanode. ZnO nano-powder with the diameter of 20-40 nm and a purity of 99.9% was purchased from Sigma Aldrich and was used as a received. N-719 industry standard dye with the molecular weight of 1,188.55 was obtained from DYESOL Ltd., Australia in a powder form. Lithium iodide anhydrous (LiI) was acquired from Sigma Aldrich and Iodine (I_2) with the purity of 99.8% was obtained from Merck KGaA and both of them are in solid form. Chloroplatinic acid hydrate was purchased from Sigma-Aldrich and was used to produce Pt paste to coat FTO glass slides to form the counter electrode.

For the DSSCs fabrication, FTO glass slides were cut to size (~2 × 2 cm^2), cleaned in an ultrasonic bath using Alconox detergent powder (Union Science, Thailand), distilled water (Union Science, Thailand), acetone (RCI Labscan Limited) and isopropanol (Union Science, Thailand), with each step requiring 30 min of treatment respectively and dried under ambient conditions inside the desiccator. ZnO paste was prepared using 2 g of Poly Ethylene Glycol 20000 (Bio Basic Inc Company) mixed with 18 ml of distilled water. Then, 5 g of ZnO nono-powder was added into the solution and the as-prepared ZnO paste was stirred for 30 min using the magnetic stirrer. The paste was then left overnight in order to obtain a thick slurry. ZnO paste was subsequently coated onto the FTO to define the active area of the photoanode with an area of 0.4×0.4 cm^2 using the screening technique. The photoanode was then annealed at 400°C for 1 hr to eliminate any trapped solvent in the ZnO paste. After that, the photoanode was coated with the dye solution using the dip coating technique. A 0.05 wt% solution of N-719 industry standard dye was obtained by dissolving the N-719 powder in 1:1 volume ratio of acetonitrile (Union Science, Thailand) mixed with tert-butanol (Union Science, Thailand). Various conditions for N-719 DSSCs fabrication were investigated including the number of ZnO layers (1, 2, 3 and 4 layers), impregnation times of (2, 6, 8 and 24 hr) and temperatures of (25, 40 and 60°C)

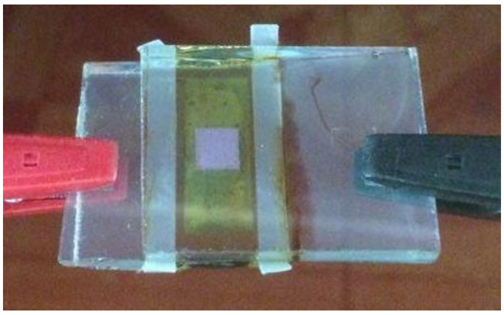

Figure 2 The complete DSSCs with the electrolyte in the middle and Para film shielding.

Pt paste was prepared using 21 mg of Chloroplatinic acid hydrate blended with 1 ml of acetone and the solution was homogenized for 30 min. The paste was screened onto FTO glass slides and was annealed at 550°C for 1 hr to complete the counter electrode preparation. The LiI/I_2 electrolyte solution was obtained using 0.27 g of LiI mixed with 0.05 g of I_2 in 10 ml of acetonitrile. The solution was stirred for 1 hr and was left overnight in the dark before it could be used in DSSCs fabrication. The DSSCs were consequently manufactured by sandwiching the photoanode and the counter electrode together with the LiI/I_2 electrolyte in the middle of the device. Finally, the DSSCs were completed by covering the device with Para film (Union Science, Thailand) and hot air from the dryer was used to provide heat in order to transform the Para film to shield the device. The complete DSSC is shown in Figure 2. The current density-voltage (J-V) characteristics of the devices were measured using a voltage-source meter (2611, Keithley Instruments, UK). An AM 1.5 solar light source (SolarLight) with an intensity of 1,000 W/m^2 was used to simulate the sunlight for efficiency measurement.

3. Results and discussion

Figure 3 shows the J-V curves of the DSSCs when the photoanode was dipped into the N-719 dye solution with dipping times of 2, 6, 8 and 24 hr at 25°C. The results showed that the open circuit voltage (V_{oc}) is unaffected by the soaking time, where the V_{oc} of the devices fabricated from each soaking time was found to be in the range of 0.56-0.6 V. The short circuit current density, on the other hand, gets affected by the soaking time and so is the performance of the device. The short circuit current density (J_{sc}) decreases from 6.66 mA/cm^2 to 4.31 mA/cm^2 when the soaking time increases from 2 hr to 24 hr. When examined closely, the J_{sc} reduces exponentially when the soaking time increases as shown in Figure 4. The reduction in J_{sc} appears to be very distinct when the coating time increases from 2 hr to 10 hr where the lowering of J_{sc} by about 20% is estimated. However, this reduction trend seems to disappear when the soaking time is more than 10 hr and the device provides a constant value of J_{sc}, at about 4.3 mA/cm^2, when the soaking time is greater than 24 hr. All important electrical parameters of the N-719 DSSCs including maximum current (I_m), maximum voltage (V_m), maximum power (P_m), fill factor (FF) and electrical conversion efficiency are displayed in table 1.

Table 1 The efficiency and the fill factor of the N-719 DSSCs fabricated using different soaking time

Time (hr)	2	6	8	24
I_{max} (mA)	0.82	0.67	0.60	0.47
V_{max} (V)	0.43	0.38	0.40	0.42
P_{max} (mW)	0.35	0.25	0.24	0.20
FF	0.55	0.49	0.50	0.47
Efficiency (%)	2.20	1.59	1.49	1.23

It is noted that I_m can be calculated by multiplying the maximum current density (J_m) with the active area of the device. The results for the I_m and V_m showed a similar trend as the J_{sc} and V_{oc} where, the current is greatly affected by the soaking time but not the voltage. The efficiency of the device fabricated with various soaking times was then calculated using maximum output power and was found to be in the range of 1.2-2.2%. The maximum cell efficiency of 2.2% as well as the maximum fill factor of 0.55 is achieved with 2 hr of soaking time. It was observed that the efficiency of the N-719 DSSCs reduces with increased soaking time and the lowest efficiency of 1.2% was measured from the device fabricated with 24 hr of soaking time. Rahman et al. have studied the effect of dipping time where the N719/ZnO DSSCs are prepared with the dipping time between 0.5-12 hr [13]. They have shown that the efficiency of the device varies slightly with the dipping time but the efficiency of the device, about 0.114-0.135%, is a lot lower compared to our study.

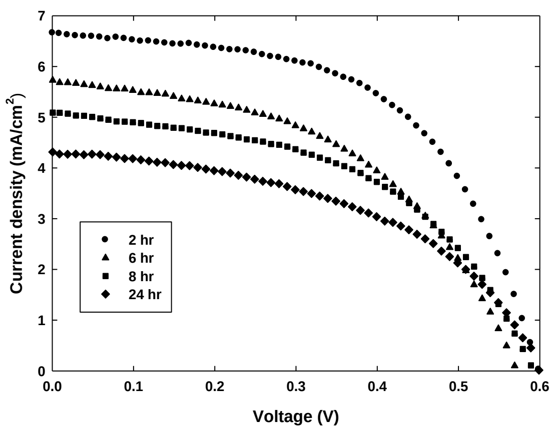

Figure 3 Current density-voltage (J-V) characteristics for DSSCs fabricated using different soaking periods

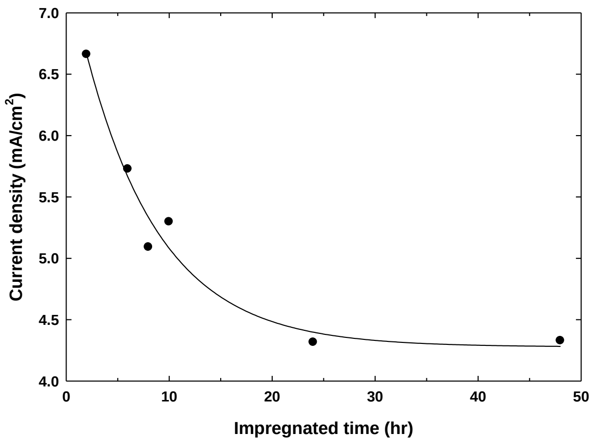

Figure 4 short circuit current density of the N-719/ZnO DSSCs as a function of soaking time D or C

Two processes that control the amount of generated current in our device are electron-hole pair separation at the N719/ZnO and free carrier transportation. The N-719/ZnO interface plays a key role in free charge carrier generation. The electrons in N-719 dye molecules are excited by the incident sunlight and the charge carriers will consequently be generated. These carriers must be quickly separated at the N-719/ZnO interface in order to become free and must be transported to the respective electrode otherwise the free carrier will be lost and lower generated current will be obtained. The ability to transport the free carrier reduces when the formation of N-719/N-719 increase. The role of the dye molecule is to absorb light and generate an electron-hole pair and not to transport the free carrier. Prolonged immersing of the photoanode in the dye solution might enhance the formation of N-719/N-719 dye molecule interfaces which could lower the capability of the device to transport free charge carriers. Low charge transport leads to the reduction of maximum current and hence diminishes the efficiency of the device.

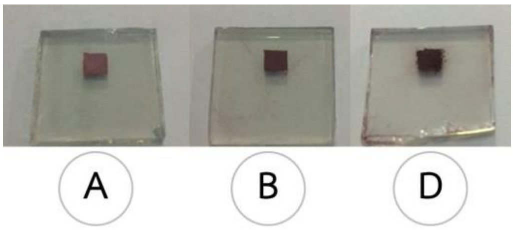

Figure 5 The color tone of the photoanode surface when impregnated in the N-719 dye solution at (a) 25°C, (b) 40°C and (c) 60°C

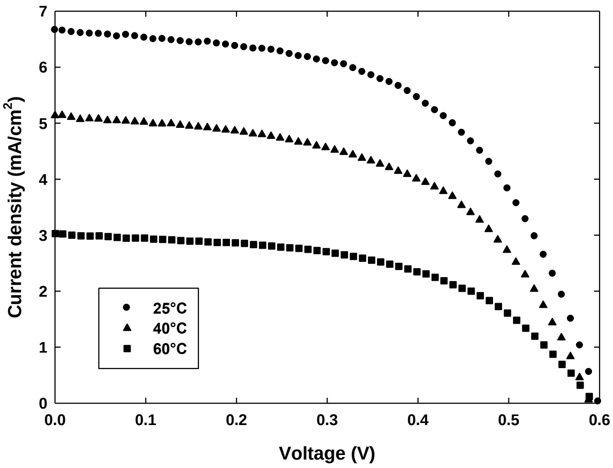

Figure 6 Current density-voltage (J-V) characteristics for N-719 DSSCs fabricated using different impregnation temperatures.

The effect of impregnation temperature during the fabrication of the photoanode was examined. Figure 5a-c reveals the difference in color tones of the photoanode when impregnated in the N-719 dye solution at temperatures of 25°C, 40°C and 60°C for 2 hr. It was seen that higher impregnation temperature produced darker tones for the N-719 dye layer on the ZnO surface. While the impregnation time obviously affects the N-719 DSSCs, the temperature during the coating process also plays a vital role on the performance of the device. Figure 6 demonstrates the current density-voltage (J-V) curves of the N-719 DSSCs for the coating temperatures of 25°C, 40°C and 60°C. At 25°C, the device provides the highest short circuit current density of 6.66 mA/cm^2 which is 55% higher than that at 60°C. On the contrary, the DSSCs fabricated at various impregnation temperatures show a constant open circuit voltage 0.6 V.

Table 2 The efficiency and the fill factor of the N-719 DSSCs fabricated using various impregnated temperatures

Temperature (°C)	25	40	60
I_{max} (mA)	0.82	0.61	0.37
V_{max}(V)	0.43	0.43	0.41
P_{max} (mW)	0.35	0.26	0.15
FF	0.55	0.54	0.53
Efficiency (%)	2.20	1.62	0.94

Table 2 shows all relevant solar cell parameters. It is noticeable from the table that the maximum voltage is unaffected by the impregnation temperature but the maximum current demonstrates a similar reduction as observed in the J_{sc}. The calculated efficiency of the N-719 DSSCs is between 0.94-2.20% where the maximum efficiency occurs at the impregnation temperature of 25°C. The fill factor, however, is relatively stable considering the coating temperatures and the values are determined to be 0.53-0.55. The result in table 2 show explicitly that the temperature during the impregnation of the photoanode in the N-719 dye solution affects the performance of the DSSCs. Under the ambient condition, coating a ZnO photoanode with N-719 dye solution at higher temperatures poses a serious threat to the quality of the dye molecules as they can be degraded at high temperature due to the reaction with oxygen and water from the environment [14]. Jo et al. also found a similar reduction of N719/TiO_2 DSSCs efficiency when the photoanode is prepared at a higher soaking temperature [15].

Figure 7 The ZnO surface structures when coated with (a) 1 layer, (b) 2 layers, (c) 3 layers and (d) 4 layers.

In the last experiment, the number of ZnO layers on the photoanode side was tested to determine the effect on performance for the device. In this experiment, 1, 2, 3 and 4 layers of ZnO were casted and baked at 400°C in the furnace before being coated with the N-719 dye solution. Figure 7 reveals the characteristics of the ZnO surface fabricated using 1, 2, 3 and 4 layers. It can be seen from the photograph that 1 layer of ZnO provides the best surface topography (Figure 7 (a)) and the quality of the surface topography declines with increasing the number of ZnO layers (Figure 7 (b)-(d)).

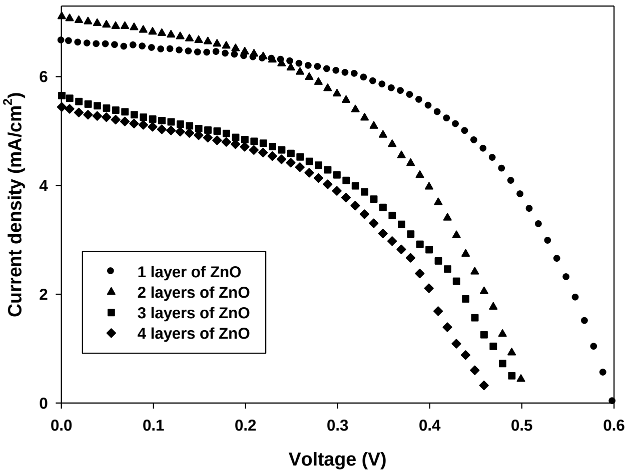

Figure 8 Current density-voltage (J-V) characteristics for N-719 DSSCs fabricated using different number of ZnO layers.

Figure 8 presents the current density-voltage (J-V) curves of the DSSCs under the illumination of 1,000 W/m^2 of sunlight from the solar simulator, when the device consists of a different number of ZnO semiconductor layers. In this case, the voltage of the device is found to be directly affected by the number of ZnO coating layers. It was seen that a single layer of ZnO semiconductor provides the maximum value of V_{oc} of 0.6 V. As the number of ZnO layer increases, however, the V_{oc} decreases. The lowest value of 0.46 V (23% lower) is extracted from the J-V curve for the device with 4 layers of ZnO.

Table 3 The efficiency and the fill factor of the N-719 DSSCs fabricated using different number of ZnO layers and temperatures

Number of ZnO layers	1	2	3	4
I_{max} (mA)	0.82	0.81	0.62	0.64
V_{max} (V)	0.43	0.34	0.33	0.29
P_{max} (mW)	0.35	0.28	0.20	0.19
FF	0.55	0.49	0.46	0.47
Efficiency (%)	2.20	1.73	1.28	1.17

Table 3 illustrates all relevant solar cell parameters that were obtained from the J-V curves. It can be seen that the maximum currents, 0.82 mA and 0.81 mA respectively, are very similar for the devices with 1 and 2 layers of ZnO. But when the number of ZnO layers increases to 3 or 4 layers the maximum current drops by about 24%. The maximum voltage has a similar reduction as in the V_{oc} when the number of ZnO increases. The calculated efficiency of the N-719 DSSCs fabricated using various numbers of ZnO layers is found to be between 1.17-2.20% where the maximum efficiency occurs when the device consists of a single layer of ZnO. The fill factor also exhibits a similar trend where the DSSC with the highest fill factor is acquired with a single layer of ZnO. The performance of the devices is clearly affected by the morphology of the ZnO surface. Increasing crack formation reduces both N719/ZnO interfacial area and the charge transport ability. The reduction of N719/ZnO interfacial area affects the junction voltage and hence the reduction of open circuit and maximum voltage. While the lowering of charge transport ability results in the reduction of short circuit current and maximum current. On the contrary, Xu et al. have discovered that a ZnO bilayer film electrode consisting of ZnO micro-flowers film as a scattering layer over a ZnO nanoparticles film as an under layer can harvest more sunlight which results in higher efficiency of the N719/ZnO DSSCs [16].

4. Conclusion

The N-719/ZnO DSSCs are manufactured with a variety of fabrication conditions in order to obtain the most efficient device. It was discovered that a single layer of ZnO impregnated in the N-719 dye solution for 2 hr at 25°C provides the best condition for fabricating the DSSCs. The efficiency and the fill factor of the device fabricated using optimal conditions were calculated to be 2.2% and 0.55, respectively. The impregnation time and temperature obviously affect the generated current but not the voltage. The number of ZnO layers affects both the generated current and voltage. The efficiency of the device drops when the number of ZnO layers, the impregnation time and temperature increases. This work opens up the possibility of producing new types of solar cells that have the flexibility and ability to coat over curved surfaces.

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