Research Article

Plasmon-Enhanced Efficiency in Dye Sensitized Solar Cells Decorated with Size-Controlled Silver Nanoparticles Based on Anthocyanins as Light Harvesting Pigment

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Abstract: The enhancement in efficiency of dye sensitized solar cells modified with silver nanoparticles based on anthocyanins as light harvesting pigment through successive ionic layer adsorption and reaction (SILAR) was demonstrated. Studies indicate that, the short-circuit current density ($J_{SC}$) and open-circuit voltage ($V_{OC}$), of DSSCs containing AgNPs were significantly improved. The photovoltaic (PV) performance decreased with increasing cycle of AgNPs from one SILAR cycle to two SILAR cycles. The best device was achieved using the photoanode prepared with one SILAR cycle. An enhancement of 35.8 % was achieved when the thickness was around 16 nm (one SILAR) over the bare FTO device. When the size of AgNPs was around 32 nm (two SILAR), an enhancement of 10.4% was recorded over the reference device. This selective enhancement in efficiency in the Ag plasmonic absorption regions is indicative of the fact that the incorporation of metal nanoparticles is beneficial for enhanced absorption and charge separation.

Keywords: Nanocomposite, SPR, Intensified Near Field, DSSCs, Silver Nanoparticles, Anthocyanins

1. Introduction

Solar cell technologies have been categorized into three generations [1]. The first generation photovoltaic solar cells are based on a single crystalline semiconductor wafer. The second generation solar cells utilize thin layer of polycrystalline semiconductor, they are cheaper to produce, flexible and lightweight; however the efficiency is still lower than first generation cells. The third generation solar cell also known as Grätzel cells or dye-sensitized solar cells was successfully achieved by combination of nanostructured electrodes and efficient charge injecting dyes [2].

However, the low efficiency of conversion and stability are the major problems confronting the third generation solar cell. Facilitating means in achieving higher efficiencies are channeled in optimizing the morphology of the active photovoltaic layer and the charge transport properties of the absorber through (i) thermal annealing treatment, (ii) use of various solvents, (iii) the use of additives and (iv) the process conditions [3-10]. Plasmonic introduction in photoactive layers of DSSC, to trap or confine light inside the active layer and enhance the absorption in the semiconductor film could result to enhanced device performances [11-22] likely due to their unique electronic, optical and magnetic properties [10, 11].

This surface plasmon resonance is dependent on the shape, size and distribution of the NPs as well as on the dielectric functions of the metal and the dielectric surrounding environment [23].

In attempt to optimize the device performance and stability, several of light harvesting materials are used to improve its photovoltaic performance and investigate their properties, since they play a very important role in harvesting sunlight and transforming solar energy into electric energy.

In addition, the dye is an important part of the DSSCs,
playing a significant role in absorbing light, generating photo-stimulated carriers and injecting these carriers into the conduction band of TiO$_2$ network. Thus, the light absorption capability of the dye and how many carriers it stimulates directly affect carrier injection and DSSCs performance [14]. Therefore, enhancing dye light absorption should be an effective way to increase the conversion efficiency.

The use of natural dyes have been considered as potential candidates to enhance the light response of semiconductor in active layers of solar cells, and have been demonstrated on several solar-cell materials [24-33].

Here anthocyanins pigment present in Hibiscus Sabdariffa extract was considered as the sensitizer due to its light harvesting capability in the visible region of the electromagnetic spectrum. Hibiscus Sabdariffa extract is rich in anthocyanins. It was reported that anthocyanin obtained from Roselle are delphinidin and cyanidin complexes [34, 35]. The chemical structure of cyanidin and delphinidin in the Hibiscus Sabdariffa dye is shown in Figure 1 [31]. It is characterized by the presence of (-OH) functional group which allows short distance between the dye skeleton and the point connected to TiO$_2$ surface, thus enhancing the binding interaction between the pigment and the TiO$_2$ film, which brings about good electronic coupling and electron transfer reaction in the visible spectrum.

![Figure 1. Chemical structures of: cyanidin and delphinidin in rosella dye.](Image)

In this present work the performance of DSSC, based on anthocyanins as light harvesting pigment when size-controlled silver nanoparticles (NPs) are incorporated into the TiO$_2$ mesoporous layer of the device structure was compared. Inclusion of AgNPs into the photoanode revealed better overall performance with an increased efficiency of 35.8% with 16 nm size. When the size of Ag-NPs was around 32 nm size, an improvement of 10.4% in efficiency was demonstrated over the performance of the reference cell. The flowers of Hibiscus were air dried till they became invariant in weight. The dried flowers of Hibiscus were left uncruushed because previous attempts proved failure to extract the dye from crushed samples due to jellification [32]. The method of heating in water was used to extract the dye. Distilled water was the solvent for aqueous extraction. 5 g of the sample (Dried Hibiscus Sabdariffa) was measured using analytical scale and dipped in 50 ml of the solvent heated to 100°C for 30 min after which solid residues were filtered out to obtain clear dye solutions.

2.2. Dye Extraction

The flowers of Hibiscus Sabdariffa were air dried till they became invariant in weight. The dried flowers of Hibiscus were left uncruushed because previous attempts proved failure to extract the dye from crushed samples due to jellification [32]. The method of heating in water was used to extract the dye. Distilled water was the solvent for aqueous extraction. 5 g of the sample (Dried Hibiscus Sabdariffa) was measured using analytical scale and dipped in 50 ml of the solvent heated to 100°C for 30 min after which solid residues were filtered out to obtain clear dye solutions.

2.3. Synthesis of Nanocomposite Material

Dip coating method was used to synthesize the Ag nanocomposite on the glass substrate. The microscope slide was cleaned with sodium lauryl sulphate (SLES) solution and then rinsed with deionized water three times. Thereafter dipped into a beaker containing a mixture of 2 moles concentrated Tetraoxosulphate (iv) acid and 2 moles chromic acid to make the surface hydrophilic for 10 minutes, the sample was thereafter rinsed with distilled water. After making it hydrophilic, it was immersed in 2 moles Tin chloride (SnCl$_2$) for 2 minutes then rinsed with distilled water for 2 minutes, then immersed in 0.35 mole silver nitrate (AgNO$_3$) for 2 minutes and rinsed with mixture of 150 ml distilled water (H$_2$O) and 0.4 moles hydrochloric acid (HCl) for 15 seconds. This procedure is called one cycle. It was repeated for two cycles.

2.4. Fabrication of the DSSCs

The sample of TiO$_2$ was prepared by the sol-gel technique in which 2 g of P25 TiO$_2$ powder was dissolved in 10 ml of deionized water mixed with 0.2 ml of Triton-X 100 and 0.4 ml of acetaldehyde, then vibrated ultrasonically using magnetic stirrer hotplate for 24 hours. The silver modified DSSCs were fabricated on FTO glass with the structure of FTO/TiO$_2$/AgNPs with different SILAR cycles. As reference sample, the device with architecture of FTO/TiO$_2$ was fabricated. The FTO glass substrate was first cleaned with deionized water and ultrasonicated in isopropanol for about 15 minutes. Motivated by this analysis, we utilized SILAR and screen printing method to achieve the design of FTO/TiO$_2$/AgNPs. To create the particles, a 16 and 32 nm layer of the silver metal film was deposited onto TiO$_2$ through successive ionic layer adsorption and reaction and annealed at 450°C for 30 minutes. TiCl$_4$ (40 mmoles) was spin coated at 2500 rounds/minute for 30 seconds and then sintered at 450°C for 30 minutes. To ensure the silver is protected, we treated it with 40 mmoles solution of TiCl$_4$ prepared at 40°C, then raised the temperature to 70°C and...
finally annealed at 300°C to 350°C. SiO$_2$ was deposited on the AgNPs using SILAR method with sodium silicate (Na$_2$O(SiO$_3$)) as the precursor. The minimum number of cycles that gave the cells stability was five cycles. The first cell with one Ag cycle had five cycles of SiO$_2$, and the second cell with two AgNPs cycles had ten cycles of SiO$_2$. The counter electrode was prepared by screen printing a platinum catalyst gel coating onto the FTO glass. It was then dried at 100°C and heated at 400°C for 30 minutes.

The cells were assembled by pressing the sensitized photoanode against the platinum-coated counter electrodes slightly offset to each other to enable electrical connection to the conductive side of the electrodes. Between the electrodes, a 50 µm space was retained using two layers of a thermotat hot melt sealing foil. Sealing was done by keeping the structure in a hot-pressed at 100°C for 1 minute. The liquid electrolyte constituted by 50 mM of tri-iodide/iodide in acetonitrile was introduced by injection into the cell gap through a channel previously fabricated at opposite sides of the hot melt adhesive, the channel was then sealed.

2.5. Characterization and Measurement

The current density-voltage (J-V) characteristics of the cells were recorded under an irradiance of 100 mW/cm$^2$ (AM1.5) simulated illumination (Keithley 2400 source meter from a Newport A solar simulator). The surface morphologies of the TiO$_2$ and TiO$_2$ with different SILAR cycles of AgNPs photoanodes were observed by scanning electron microscopy (Phenom Pro X model, Eindhoven de Netherlands). The absorption spectra of the dye and various photoanode were recorded on Ava-spec-2048 spectrophotometer in the region of 350–1000 nm. The effective irradiated area of each cell was 1.8 cm$^2$. Thickness measurement was obtained with a Dektac 150 surface profiler.

3. Results and Discussion

Figure 2 depicts a representative ultraviolet (UV)-visible (vis) absorption spectra of *Hibiscus Sabdariffa* extract. The absorption peak of dye extract was observed around 550 nm (indicated in Figure 2) which ascertains the presence of anthocyanin pigment and in agreement with Onimisi et al [13].

![Figure 2. UV-Vis spectra of the dye.](image)

Fig 3 shows the UV-vis absorption spectra of the TiO$_2$ and TiO$_2$ with different SILAR cycles of AgNPs within the wavelength range of 350-1000 nm in the absence of anthocyanin pigment along with the controlled size growth of the Ag particle from 16 to 32 nm which correspond to the number of cycles as shown in Figure 5.

![Figure 3. UV-vis spectra of various prepared Ag NPs suspensions without dye.](image)
Figure 4. UV-vis spectra of various prepared Ag NPs suspensions with dye.

Figure 5. Thickness measurement of 10 SILAR cycles which shows the growth rate of 1 and 2 cycles.
Fig 4 shows the UV-vis absorption spectra of TiO$_2$ and TiO$_2$ with size controlled AgNPs after sensitization. The absorption of the entire visible region for the electrode with different SILAR cycle of AgNPs was stronger than that for the electrode without AgNPs. The optical absorption enhancement was observed in the dye-loaded plasmonic nanocomposite films which is attributed to the SPR of metallic AgNPs, which interacted with the dye molecule thereby enhancing dye absorption.

These properties exhibited above shows that dye molecules in the presence of metallic NPs can absorb more photons, possibly because of the intensified near-field effect of the surface plasmon and spectral overlap between the dye and SPR.

Figure 6. SEM images of (a) 0 SILAR cycle, (b) 1 SILAR cycles and (c) 2 cycles.
The performance of the DSSCs based on only TiO$_2$ and Ag modified TiO$_2$ electrodes with one and two SILAR cycles were examined under 1 sun AM 1.5 simulated sunlight. Figure 7 and Table 1 compared the $J$–$V$ characteristics for DSSCs prepared with the TiO$_2$–Ag with 1 and 2 cycles nanocomposite and TiO$_2$ NPs electrodes with an effective area of 1.80 cm$^2$. As demonstrated from Table 1, the photovoltaic parameters, especially the photocurrent, and open circuit voltage of DSSCs containing Ag NPs was affected by the SILAR cycle of AgNPs. The PV performance decreased with increasing size of AgNPs from 16 nm to 32 nm. From Table 1, the best performance was achieved using the device fabricated with 16 nm size of AgNPs. The best cell demonstrated a $J_{sc}$ of 0.0385 mA/cm$^2$, $V_o$ of 0.45 V and $FF$ of 0.526, yielding the highest efficiency ($\eta$) of 0.0991%. The cell exhibits a 35.8% improvement over the performance of bare FTO-based device. The improvement in efficiency in the presence of Ag NPs can be seen to arise from an increase in $J_{sc}$ and $V_{oc}$ values. The enhanced $J_{sc}$ is related to the light harvesting capability of dye molecules by excitation of the SPR.

It is shown from Figure 7, that the mixed films containing Ag particles exhibit an increase in the power conversion efficiency from 10.4 to 35.8%.

Table 1. Performance characteristics of DSSCs fabricated with different anodes under 100 mWcm$^2$.

| Sample | $J_{sc}$ (mAcm$^{-2}$) | $V_{oc}$ (V) | $FF$ (%) | $\eta$ (%) |
|--------|-----------------------|--------------|----------|------------|
| FTO/TiO$_2$ | 0.026 | 0.432 | 59.4 | 0.0067 |
| FTO/TiO$_2$/AgNPs (1 SILAR) | 0.039 | 0.450 | 52.6 | 0.0091 |
| FTO/TiO$_2$/AgNPs (2 SILAR) | 0.034 | 0.449 | 48.8 | 0.0074 |

The improvement in efficiency in the presence of Ag NPs can be seen to arise from an increase in the power conversion efficiency from 10.4 to 35.8%.
As shown in Table 1 above, for better performance, Metal plasmon must be 16 nm in size to escape recombination and produce an enhanced photovoltaic parameter. The electric field of localized plasmon resonances only promotes few dye molecules when the AgNPs size was 32 nm, leading to limited enhancement. Therefore, light trapping of localized surface plasmon was mainly improved in DSSC fabricated with FTO/TiO$_2$/AgNPs (16 nm) photoanode electrode in our system, leading to the enhanced photocurrent response in the whole visible region.

4. Conclusions

The impact of size-controlled AgNPs on the performance of DSSCs through successive ionic layer adsorption and reaction based on anthocyanins pigment was demonstrated. The photovoltaic performance was evaluated under 100 mW cm$^{-2}$ light intensity. The effect was studied in the system of modified size-controlled Ag inclusions inside the DSSC. The photovoltaic response of the nanocomposite with inhomogeneous distribution of coated two dimensional array cluster of NPs randomly dispersed on the top surface of active TiO$_2$ nanoporous layer was analyzed.

The performance, especially the photocurrent, and open circuit voltage of the DSSC containing AgNPs was affected by the controlled sizes of AgNPs. The modified AgNPs photoanode with 16 nm thickness shows an efficiency of 0.0091% which represents a 35.8% improvement in efficiency over the DSSC without AgNPs. Also when the size of the AgNPs was 32 nm, an efficiency of 0.0074% was achieved which represents a 10.4% improvement from the standard electrode without AgNPs.

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