



# A Study on the Effect of Gold Nanoparticles for Dye Sensitized Solar Cell Using Hibiscus Rosa-Sinensis as Photosensitizer

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**Abstract:** Dye-sensitized solar cells (DSSC) mainly use organic components as an alternative way of light harvesting element to replace the traditional silicon solar cells. Natural dye alternatively is being used as sensitizers to overcome the limitation of inorganic ruthenium dye which contains heavy metal. In this study, DSSC is expected to enhance its optical properties and light absorption by implementing gold (Au) nanoparticles. Various natural and organic sources can be extracted and acted as a photosensitizer to trap solar energy. Fresh Hibiscus Rosa-Sinensis flowers are one of the good candidates to give higher light absorption in DSSC. The characteristics of the dye photosensitizer will be observed under UV-Vis-NIR Spectrophotometer. The efficiency of complete DSSC will be determined by using LIV tester. An attempt to enhance light-harvesting efficiency and hence the light to current conversion efficiency by mixing Au nanoparticles TiO<sub>2</sub> paste. Au nanoparticle was selected because it can act as a medium between the dye and TiO<sub>2</sub> to increase the electron transmission speed. The Hibiscus dye in DSSC achieved an efficiency of 0.14%. However, the efficiency reduced to 0.000178% as Au NPs was added. The effects of the addition of Au nanoparticles are discussed.

**Keywords:** Dye-sensitized solar cells, Hibiscus rosa-sinensis, gold nanoparticles, photosensitizer, efficiency

## 1. Introduction

Natural phenomena always become an idea for human to invent a new concept to make human life easier. One of the notable examples is photosynthesis, that convert light energy into chemical energy. In 1974, similar concept of photosynthesis was introduced by Melvin Calvin that convert light into electrical power [1]. Then, 14 years later, a dye-sensitized solar cell (DSSC) was invented by Brian O'Regan and Michael Grätzel which based on a semiconductor formed between a photo-sensitized anode and an electrolyte [2].

DSSC provides a feasible alternative to the inorganic p-n junction solar cells by converting solar energy into electric energy through the photosensitization of the cell [3], [4]. It can be a solution to the inorganic materials solar cells which required expensive methods during production caused by the p-n/heterojunction or metal-semiconductor junctions. Therefore, DSSCs offers a relatively low in cost and simple method in production [5]. Furthermore, DSSCs work well under low irradiation conditions, and suitable to be place at the cloudy weather countries. Nevertheless, the

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efficiency of DSSCs still low and need to be increased in order to be compatible with others solar cell in current market. Moreover, the evaporation and leakage of the electrolyte resulted a question on its long-term stability [6].

In this research, DSSC by using abundantly available Hibiscus rosa-sinensis aimed to reduce the high cost due to the expensive methods of inorganic solar cell and metal nanoparticle is expected to help low light absorption and thus enhanced low efficiency facing by DSSC.

## 2. Literature Review

Many efforts and studies have focused on dye photosensitizer since dye plays an increasingly important key role in harvesting sunlight and converting solar energy into electrical energy. Historically, in 1991, Ruthenium-complex dyes including metal-free organic dyes and metal-complex porphyrin dyes are used to fabricate DSSC with conversion efficiencies of over 10% [2]. However, those type of dyes have already intensified due to the high cost of ruthenium itself and yet derived from relatively scare natural resources corresponds to a heavy environmental burden [7]. Hence, it is possible to use natural dyes as alternative photosensitizers with appreciable efficiencies. Their advantages over synthetic dyes include easy availability, abundance in supply, can be applied without further purification, environmentally friendly and they considerably reduce the cost of devices [8].

Additionally, importance is given to the naturally available dyes found in flowers, fruits and vegetables from which the pigments are extracted in a cost-effective manner and tested for their performance [9]. Anthocyanins, Betalain, Chlorophyll and pigments, which are generally found in the bright coloured fruits, flowers, and vegetables, are successfully used as photosensitizers in DSSCs. Therefore, anthocyanins are basically responsible for colours from red to blue range found in the leaves, flowers, fruits, stems, roots of the plant with light absorption between 300-800 nm wavelengths and they are flavoid pigments which are in abundance [10], [11], [12]. The carbonyl and hydroxyl groups found in the anthocyanins can bind themselves to the TiO<sub>2</sub> film, which relatively facilitates the electron transfer from the dye molecule into the conduction band of TiO<sub>2</sub> layer. In the previous reports, various natural source of Anthocyanin gives variety of sensitized performance [13], [14]. In this paper, the DSSC is prepared using natural flower dye extracted from Hibiscus Rosa-Sinensis. Thus, Hibiscus Rosa-Sinensis based dye has been selected due to its widely availability in local area of Malaysia. Wong et al. reported the red flower of this Hibiscus Rosa-Sinensis contains high concentrations of anthocyanins that gives higher potential of natural dyes in solar cells [3]. However, further research and studies regarding the performance of hibiscus dye-sensitized solar cells is still lacking and further investigation need to be done.

There are many attempts made to find a solution to the drawbacks of DSSCs. Recently, some research has shown that surface plasmon resonance from metallic nanostructures can improve the light absorption on photoelectrode of DSSC [15], [16]. Particularly, Gold nanoparticles (Au NPs) are a great interest due to intense absorption in the visible region which is due to the surface plasmon resonance effects [17]. Therefore, in this research, in conjunction with a natural dye extracted from hibiscus flower, Au NPs will be integrated to exploit the surface plasmon resonance phenomena in order to enhance the efficiency of the DSSC.

## 3. Methodology

### 3.1 Preparation of Conductive Fluorine-doped Tin Oxide (FTO) Glass

Clear conductive glass is commonly used as substrate because of its relatively high optical transparency in the visible and near infrared regions of the electromagnetic spectrum. Conductive coating (film) in the form of transparent conductive oxide (TCO) layer is deposited on one side of substrate. This layer is essential since it allows sunlight penetrating the cell while conducting electron carriers to outer circuit. The conductive film ensures a very low electric resistance per square. Typical value of such resistance is 10-20  $\Omega$  per square at room temperature. The nanostructure wide band gap oxide semiconductor (electron acceptor) is applied, printed, or grown on the conductive side. Meanwhile, this study utilized Fluorine-doped Tin oxide (FTO) glass as the substrate with 10  $\Omega$  of resistance value.

The preparation of the electrode started by cutting the FTO glass into a slide of 3 cm square shape using diamond glass cutter. Then, cut conductive glasses were merged into beaker containing 95wt.% ethanol solution. It then underwent ultrasonic bath for 10 minutes at medium mode for cleaning process. After that, the FTO glasses were submerged in a beaker containing distilled water and preceded to the ultrasonic bath for another 10 minutes.

### 3.2 Preparation of Semiconductor Film Photoelectrode (TiO<sub>2</sub>)

The photoelectrode or working electrode in a DSSC is made up of a nanostructure semiconductor material, attached to a transparent conducting substrate. The most extensively utilized semiconductor material is TiO<sub>2</sub> which is anatase band gap 3.2 eV [18]. Thus, the electrode consists of interconnected nanoparticles that is in size range of 15-30 nm and forms a transparent porous electrode.

Titanium (IV) oxide (TiO) aerioxide nanoparticles P25 ~21 nm primary particle size had been selected in this study. A porous TiO<sub>2</sub> paste was prepared, in which the 5g of TiO nanoparticles P25 was in nanopowder form and grinded together with the 11 mL of 0.1M of glacial acetic acid solution in a mortar and pestle under a room temperature of 30°C

with well ventilation. The mixture was well grinded for 30 minutes until it showed a result of a very light white milky – paint like solution. Then, the paste was equilibrated by adding the surfactant which contained a mixture of one drop of dishwashing detergent and 6 mL of distilled water. Extensive stirring was carried out to ensure a complete dispersion of TiO<sub>2</sub> nanoparticles with other substances and they can mix homogeneously. The surfactant acted as to reduce the surface tension of the solution in which it dissolved.

### 3.3 Preparation of Photoanodes

The conductive side of the FTO glasses was determined by checking the resistance value of both surfaces of the glass slide by using a digital multimeter. The conductive side will show a reading of resistance values while the non-conductive side will show zero value. Adhesive tapes are attached with dimension 1.5 cm<sup>2</sup> on every edge of FTO glass to give a guide to spread the pastes. A few drops of TiO<sub>2</sub> paste was applied onto the conductive side surface of the glass substrate and performed a ‘doctor blading’ technique to spread the TiO<sub>2</sub> paste evenly by using a clean glass rod. To form a well deposited thin film of TiO<sub>2</sub>, it was then placed on the turning table of a spin coater to do spin-coating for 5 minutes at 1000 rpm. The TiO<sub>2</sub> film then was annealed in the furnace at 450°C for 30 minutes. The high temperature yields in electrical interconnection between the nanoparticles and ultimately forms the nanostructure porous electrode [19]. After the annealing process, the produced electrode was then cooled down in the furnace until the temperature dropped to 50°C. This is to prevent fast cooling rate of the film slide to reduce the thermal resistance of the FTO glass.

### 3.4 Gold (Au) Nanoparticles Preparation

5 mL of gold nanoparticles with 10 nm of diameter, silica coated and dispersion in H<sub>2</sub>O was bought from Sigma Aldrich. A drop of Au nanoparticles was added to the TiO<sub>2</sub> paste by using a pipette. Then, the mixture was grinded in a mortar and pestle for 30 minutes. Extensive stirring was carried out to ensure a complete dispersion of Au NPs in the TiO<sub>2</sub> paste. The prepared Au NP - TiO<sub>2</sub> mixture was tightly capped and stored in a glass bottle.

### 3.5 Preparation of Triiodide Electrolyte Solution (I<sup>-</sup>/I<sub>3</sub><sup>-</sup>)

Short circuit current density (J<sub>sc</sub>) and open circuit voltage (V<sub>oc</sub>) considerably depend on the electrolyte. The electrolyte should also regenerate the dye efficiently after the process of electron injection to conduction band of oxide semiconductor and dye excitation. This is apart from being on electrically conducting medium in DSSC. The Triiodide electrolyte solution was prepared in a glass beaker by dissolving 5 mL of 0.5M potassium iodide (KI) and 10 mL of 0.05M of iodine solution (I<sub>2</sub>) in 10 mL of acetonitrile solvent. All solutions were mixed and stirred smoothly with a clean glass rod. Then, the electrolyte solution was tightly capped and stored.

### 3.6 Preparation of Natural Hibiscus Rosa-Sinensis Flower as Dye Photosensitizer

The fresh hibiscus flowers were collected and washed with distilled water. It was carefully dried in room temperature for a few hours and pestle properly. Then, 85 mL of acetone was poured into the dye extraction in the beaker. The solution was then stirred using a glass rod to obtain a good consistency of the dye. Finally, Whatman filter paper no 41 was used to filter the dye obtained. The dye sensitization is performed by immersing the photoelectrode into hibiscus dye solution and leaving it for 24 hours. During this stage, the colour of the TiO<sub>2</sub> layer changed from white to slightly purple. The dye-stained slide was then cleaned with distilled water to remove any excess dye on the glass. To prepare the graphite-coated slide, the conductive side of the FTO glass plate was coated with carbon by gently shading the surface with a graphite stick.

### 3.7 Assembling of Complete Dye Sensitized Solar Cell (DSSC)

Next, the cell device was assembled by placing the graphite-coated slide face down on top of the stained titanium dioxide coated side. The two opposing slides were placed on top of each other with a slight offset to ensure that all the stained titanium dioxide was covered by the counter electrode and that a space was available for connecting the crocodile clips. Next two binder clips were used on opposite edges to gently hold the slides together. Then, two drops of the electrolyte solution were placed at the edges of the plates. Subsequently, the two binder clips were alternately opened and closed to permit the uniform dispersion of electrolyte solution between the slides.

### 3.8 Characterizations

The wavelength measurement for the UV-Vis optical absorption analysis is taken within the range of 300 nm to 800 nm. The optical absorption was analyzed for the Au NPs-TiO<sub>2</sub>- Hibiscus, and bare hibiscus dye solution. The optical absorption spectrum was obtained using Optical Spectrophotometer Jasco V-670 EX. The current-voltage curves of the DSSCs were obtained by applying an external bias to the cell and measuring the generated photocurrent under white light radiation with a Light Current-Voltage (LIV) tester Keithly digital source meter. The intensity of the incident light was 100 mW/cm<sup>2</sup> and the instrument was equipped with a 300 W solar simulator.

## 4. Results and Discussion

### 4.1 Optical Characterization

Fig. 1 shows the UV-Visible absorption spectra of the fresh hibiscus dye extraction between 350 nm to 1000 nm. The of the hibiscus dye starts to incline from 350 nm to the first peak of 410 nm and declines before it increases rapidly towards the second peak at 550 nm with its tail desorbs to broader wavelength. The absorption spectrum was slightly different from those reported by Rachel and Gobinat for Hibiscus rosa-sinensis dye that was extracted using 95% ethanol solution, which resulted in an absorption peak at 516 nm [18]. This is expected due to the absorption characteristics resulted from the different types of functional groups on the anthocyanins and the colours of the extracts [18]. Absorption with different wavelength value will assist the cells to capture photons at two different energies. The resultant absorbance depends on the anthocyanin colour contained in the dye. The anthocyanins' molecular structure found in fresh Hibiscus rosa-sinensis has a good light absorption as photosensitizer in DSSC application.

Based on Fig. 2, the maximum absorption spectrum achieved by the TiO<sub>2</sub> coated with Hibiscus dye solution lies at 600 nm and slowly decreased at longer wavelength. The same tendency with higher absorption could be observed for the Au NPs doped TiO<sub>2</sub> with Hibiscus dye extraction, where the peak absorption spectra occur at 620 nm. Moreover, the Au NPs-TiO<sub>2</sub>-Hibiscus Dye shows a higher absorption compared to the TiO<sub>2</sub> - Hibiscus at longer wavelength which is favorable to the DSSC because more conversion from energy to electricity will be generated if the absorbed spectrum is wider [19].

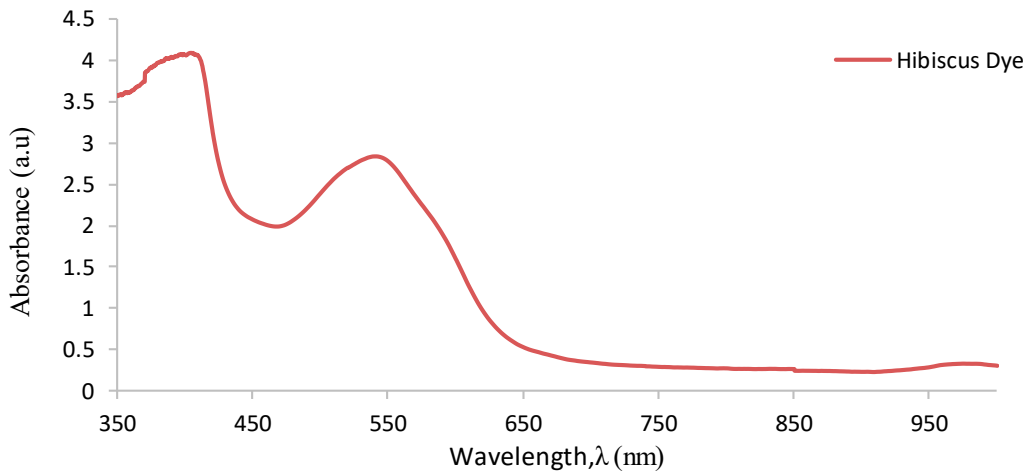


Fig. 1 - Absorption spectrum of Hibiscus Rosa-Sinensis dye extraction

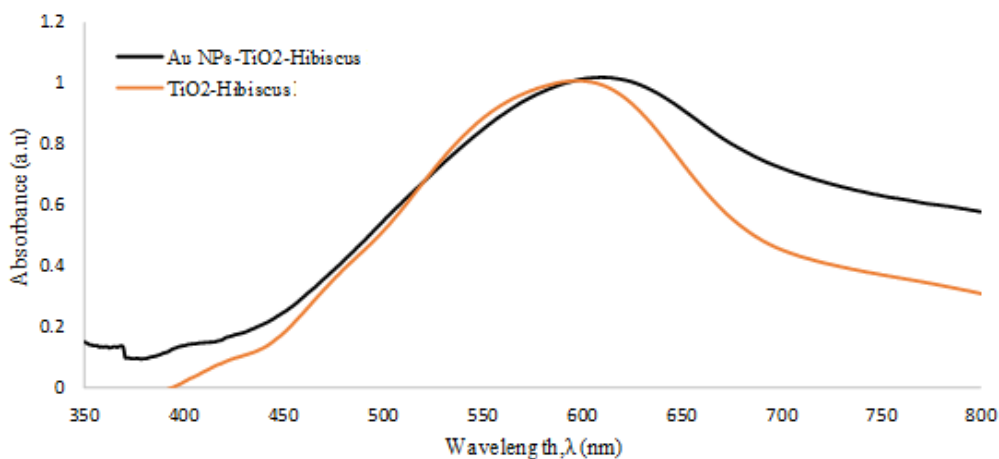


Fig. 2 - Absorption spectrum of Hibiscus Rosa-Sinensis dye extraction

### 4.2 Efficiency of DSSC

Electrical Current-Voltage (I-V) measurements of the DSSCs were measured with a Keithley semiconductor analyser using a 100 mW/cm<sup>2</sup>. Table 1 shows a clear glimpse of the photovoltaics parameters of DSSCs based on different dye configuration. The recorded open circuit voltage for natural Hibiscus dye in complete cell was 0.330 V

and the recorded short circuit current was 1.3 mA/cm<sup>2</sup>. These characteristics exhibited a fill factor of 32.44 and efficiency of 0.14%. An efficiency improvement of 27% can be observed than by reported Rachel and Gobinat et al. [18]. Acetone was a polar aprotic solvent. Acetone was found to be more effective for dye diffusion whereas ethanol being polar protic in nature was not suitable for dye diffusion which maybe the reason of the higher efficiency of Hibiscus dye DSSC in this study [20].

In addition, the higher performance resulted from a smaller particles size of the Aeroxide P25 TiO<sub>2</sub>. The nanoparticles of the Aeroxide P25 TiO<sub>2</sub> are potentially packed more efficiently onto the FTO substrate surface because the voids among the particles decrease with decreasing particle size [19]. The small size of the TiO<sub>2</sub> nanoparticles results in a large surface area that permits for maximum adsorption of the dye sensitizer on the semiconductor. Thus, this structure could offer the desired directionality for electron transport in TiO<sub>2</sub> electrode which could give a result in faster photo-response and higher electron collection efficiencies [21]. A greater number of pores in the TiO<sub>2</sub> film enhance the regeneration of the oxidised dye and the charge transport. According to Xu et al., an increase in the redox mediator transport rate in the higher porosity TiO<sub>2</sub> film was greatly enhanced in the porous structure, and thus enhanced its efficiency [22].

**Table 1 - Photovoltaics parameters of DSSCs based on different photoelectrode configuration**

Photoelectrode Type	$J_{sc}$ (mA/cm <sup>2</sup> )	$V_{oc}$ (V)	$FF$	$\eta$ (%)
TiO <sub>2</sub> - Hibiscus	1.3	0.330	32.44	0.14
TiO <sub>2</sub> - Au NPs - Hibiscus	0.1	0.006	29.71	0.00017
TiO <sub>2</sub> - N719	3.3	0.661	29.41	0.64

The role of gold nanoparticles is to act as a medium of plasmon resonance effect between the dye and TiO<sub>2</sub>, to increase the electron transmission speed and the maximum voltage. Theoretically, gold nanoparticles Au NPs are adsorbed on the surface of TiO<sub>2</sub> shift the Fermi level, allowing the electrons from the dye to be excited with low energy consumption, resulting in faster electron transfer and an improvement in the overall conversion efficiency of the DSSC [23]. However, as observed in Table 1, a substantial decrease in photocurrent output by adding Au NPs to the anthocyanin Hibiscus sensitized solar cell. It likely that resistance within the cell increased and more obstacles for the electrolyte to pass in order to recharge the anthocyanin molecules in photo-voltage which similar to the study carried out by Hailey et al., where the open circuit photovoltage was reduced due to the addition of Au NPs into blackberry anthocyanin dye [23]. Hailey et al. also suggested that anthocyanin dye and Au NPs could be allowed to absorb into the TiO<sub>2</sub> DSSC for longer periods of time and at higher concentrations to allow maximal NPs loading. A decrease in efficiency by addition of Au NPs to the photoelectrode was probably due to the shadowing and blocking of TiO<sub>2</sub> active sites by bigger and scarcely gold nanoparticles [23].

According to Table 1, the commercialized dye, N719 yielded an efficiency of 0.64% in DSSC. N719 was used as reference because of its excellent performance in terms of efficiency and stability. Dyesol's N719 industry standard dye is a modification of the N3 foundation dye with changes that increase cell voltage. Therefore, the natural dyes that contain anthocyanins molecular structure such as Hibiscus rosa-sinensis tend to have a high-performance due to their wavelengths that are almost the same with N719 which were around 500 nm to 600 nm [24]. Apart from this, by using Hibiscus rosa-sinensis dye as the photosensitizer, the performance of DSSC can be achieved from 0.6% to 1%.

## 5. Conclusions

In this study, DSSCs were prepared using dye that was extracted from the flowers of Hibiscus rosa-sinensis and the effect of gold nanoparticles onto the cells were discussed. The dye exhibited a UV-Visible absorption peak at 550 nm. The Hibiscus dye in DSSC achieved an efficiency of 0.14%. However, the efficiency reduced to 0.000178% when the Au NPs was added. The decrease in the performance of DSSC was probably due to the shadowing and blocking of TiO<sub>2</sub> active sites by bigger and scarcely effective gold nanoparticles. Nevertheless, the preparation of the Hibiscus dye as the photosensitizer for the DSSC is a promising approach, which seems to have a high-performance due to its wavelength that is almost similar with the commercialized standard dye, N719. As a recommendation, there are many aspects need to be precise or established in terms of procedure in making DSSCs.

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