RESEARCH ARTICLE

Efficiency enhancement in SnO_2 based dye-sensitized solar cells by incorporating plasmonic gold nanoparticles

K. Umair*, M.A.K.L. Dissanayake and G.K.R. Senadeera



Highlights

- Local Surface Plasmon Resonance effect by Au nanoparticles was used for the enhancement of efficiency in dyesensitized solar cells based on SnO₂.
- An impressive efficiency enhancement of more than 26% has been achieved due to the incorporation of the optimized amount of Au nanoparticles.
- Incorporation of Au nanoparticles has reduced the recombination rate of electron-hole pairs in SnO₂ photoanode.

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Efficiency enhancement in SnO₂ based dye-sensitized solar cells by incorporating plasmonic gold nanoparticles

K. Umair^{1,2,*}, M.A.K.L. Dissanayake^{1,2} and G.K.R. Senadeera^{1,3}

¹National Institute of Fundamental Studies, Kandy, Sri Lanka.

²Postgraduate Institute of Science, University of Peradeniya, Peradeniya, Sri Lanka.
 ³Department of Physics, Open University of Sri Lanka, Nawala, Nugegoda, Sri Lanka.

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Abstract: SnO₂ is an attractive semiconducting material which can be used in place of TiO₂ in dye-sensitized solar cells (DSSCs) due to the wide energy band gap, good photostability and high charge carrier mobility. Here we report the use of plasmon resonance effect by gold nanoparticles of size 70-80 nm for efficiency enhancement in solar cells made with SnO₂ photoanodes and sensitized with Indoline D149 dye. Devices were characterized by current density-voltage (J-V) curves, incident photon-toelectron conversion efficiency (IPCE) spectroscopy and electron impedance spectroscopy (EIS). DSSCs fabricated with pristine SnO₂ photoanode showed efficiency of (η) 2.28%, under the illumination of 100 mW cm⁻² (AM 1.5), whereas DSSCs fabricated with optimized Au nanoparticles in the SnO₂ photoanode showed efficiency of 2.89% having more than 26% enhancement. This increase is mainly attributed to the 42% increase in the short circuit current density from 6.48 mA cm⁻² to 9.19 mA cm⁻² caused by the plasmonic effect by Au nanoparticles. The IPCE was also increased due to the use of Au@SnO2. According to the EIS analysis, the incorporation of plasmonic Au nanoparticles has led to a 35% decrease in the interfacial charge transfer resistance (R_{CT2}) at the SnO₂ photoanode/electrolyte interface which is associated with the increased rate of charge transfer at this interface and increased resulting the efficiency of the solar cells.

Keywords: Au nanoparticles; Indoline D149; Plasmonic effect; Power conversion efficiency; SnO₂.

INTRODUCTION

Among the semiconductors which can be employed in dye sensitized solar cells (DSSCs), tin oxide (SnO₂ is one of the promising materials due to its desired properties, such as chemical stability and flexible morphological transformations with a wide band gap (E_g 3.62 eV at 27 °C), notable photo stability and high charge mobility (~250 cm²V⁻¹s⁻¹)(Barry *et a.l.*,2004; Brikel, 2012; Zainudeen *et al.*, 2019). However, due to the low power efficiencies of the DSSCs fabricated with SnO₂ as the photoanode material, little attention has been paid as compared with the TiO₂ in these devices (Barry *et al.*, 2004; Birkel *et al.*, 2012). However, different approaches have been attempted to enhance the efficiencies of DSSCs with SnO₂. Among these techniques, covering the SnO₂ crystallites with either semiconducting material such as ZnO or with insulating materials such as MgO has attracted much interest during the last couple of decades (Barry et al., 2004; Hägglund et al., 2008; Birkel et al., 2012; Zainudeen et al., 2019). Apart from these, as widely used in the TiO, based DSSCs incorporation noble metal nano particles, such as Au or Ag, which shows the Local Surface Plasmon Resonance (LSPR) effect could be another technique to enhance the efficiency of SnO₂ based DSSCs. However, according to our knowledge, no comparison has been done so far in the efficiencies of the DSSCs with and without the LSPR effect in the photoanode. These noble metal nanoparticles (NP) have been used to enhance the light absorption, and increase the charge transfer rate and the photocurrent in DSSCs. The Au NPs exhibit unique optical properties caused by the local plasmon resonance effect at the NP surface (Hägglund et al, 2008; Zainudeen et al, 2019). At a wavelength of light, characteristics of the type and size of the metal NPs, collective oscillations of electrons occur at the nanoparticle surface resulting in the LSPR or plasmonic effect causing strong resonance absorption and scattering of light. This LSPR effect makes the electromagnetic fields near the metal NPs stronger, and creates a strong optical absorption band in the UV-vissible region of the spectrum. The improvement in efficiency is attributed to a higher value of photocurrent in the plasmonic devices arisen due to near-field enhancement effects. The plasmonic resonance effect by enhanced light absorption, therefore, enhances the absorption of light by the sensitizing dye for improved efficiency in solar cells (Hägglund et al., 2008; Jeong et al., 2011 Zainudeen et al., 2019). According to the details published by Ariyasinghe et al, Indoline D149 dye is the most suitable dye for the SnO₂ based DSSCs. The passivation of trap sites in the SnO₂ film attributed to the good performance of SnO₂ based DSSCs than ruthernium dye (Ariyasinghe et al., 2011). By considering these facts, the possibility of using the LSPR effect in Indoline D149 sensitized SnO₂ photoanodes towards the enhancement of efficiency in DSSCs fabricated with SnO₂ photoanode is reported for the first time.



(D https://orcid.org/0000-0002-5566-5275

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MATERIALS AND METHODS

Preparation of gold nanoparticle suspensions

AuNPs were prepared by the citrate reduction method. Hydrogen tetrachloroaurate(III) (HAuCl₄, Fluka) and trisodium citrate (Na₃C₆H₅O₇.2H₂O) (Sigma–Aldrich) were used as starting materials. A 20.00 mL solution of 1.0 mmol L⁻¹ HAuCl₄ was prepared by adding deionized water, and the solution was stirred under boiling. A 2.0 mL of 1% Na₃C₆H₅O₇ solution was quickly poured into the solution of 1.0 mmol L⁻¹ HAuCl₄. It was observed that the solution turned from blue to reddish–pink. Figure 1 shows the colour of the gold nanoparticle solution prepared by rapid citrate reduction method. It was reddish–pink colour with large particle size (70–80 nm) (Njoki *et al.*, 2007; Tabrizi *et al.*, 2009).



Figure 1: Colloidal gold nanoparticles of 70-80 nm size range.

Optical absorption of Au NP colloidal solutions

A Shimadzu 2450 UV–Vis spectrophotometer was used to record the optical absorption spectrum of the Au NP colloidal solutions from 200 to 1100 nm wavelength region. Gold nanoparticles of different sizes have specific absorption spectra. When the diameters of nanoparticles are between 12 and 80 nm, the maximum absorption peaks are located at 520–550 nm, and there are red shifts observed gradually with the increase in diameters of gold nanoparticles (He *et al.*, 200). For the Indoline D149 dye, the time dependent density functional theory calculations revealed that there were several kinds of conformations resulting in two types of absorption spectra. The first type showed a long-wavelength region around 530 nm, whereas the second type showed a short-wavelength region around 390 nm (Hiroaki *et al.*, 2013).

Measurement of particle size of plasmonic Au NP

The particle size of Au NP colloidal solutions was measured by HORIBA nanoPartica SZ-100V2 instrument. Particle size analysis was performed by dynamic light scattering (DLS) method (Scattering angle, 90°) and the dynamic range was 0.3 nm–8 μ m.

Preparation of SnO_2 photoanode and J-V and EIS characterization

Fabrication of bare SnO_2 films was done as follows: SnO_2 colloidal solution (Alfa chemicals, 15% SnO_2 colloidal in H₂O) was ground together with 1.0 mL of glacial acetic

acid and a few drops of Triton–X 100 using an agate mortar. The mixture was dispersed in 40.0 mL of ethanol and ultrasonicated for 15 min. The dispersion was sprayed onto fluorine doped tin oxide (FTO) glass substrates (sheet resistance 8 W /sq) heated at 150 °C, and then sintered at 550 °C in a furnace for 45 min. The area of the FTO substrates was kept as 1.0 cm \times 2.0 cm.

SnO₂ photoanodes (AuNP@SnO₂) incorporated with Au nanoparticles were fabricated by solution drop casting method while keeping the FTO substrates with SnO, films at 100 °C on a hot plate. Different amounts of AuNP colloidal suspension (by volume) were drop casted on the above FTO/SnO2 substrates. The area of solar cells was maintained at 0.25 cm², and the thicknesses of all the SnO₂/FTO films were kept constant throughout the investigation. Thin films of SnO₂ on the FTO with and without Au nanoparticles were then immersed in separate 0.3 mmol L⁻¹ Indoline D149 dye solutions (in t-butyl alcohol : Acetonitrile, 1:1 by volume) for 1 h and 45 min and gently rinsed with absolute ethanol. The liquid electrolyte containing I^{-}/I_{3}^{-} redox mediator thus prepared was injected into the space between the photoanode and the counter electrode to form a sandwich structure for each photoanode type. Current-voltage measurements of each cell was obtained under the simulated sun light of 100 mW cm⁻² with AM 1.5 spectral filters. Optimized amount of Au nanoparticles were determined by measuring the efficiency of each solar cell fabricated with different amounts of Au. Comparison of the efficiencies of both the devices fabricated with and without Ag nanoparticles was done under the same conditions. Electrical impedance spectroscopic (EIS) measurements were carried out in the frequency range between 0.01 Hz-1.0 MHz by using the Autolab Potentiostat/ Galvanostat PGSTAT128 N under the same illumination as above. (Aponsu et al., 2010; Dissanayake et al., 2016).

IPCE measurements

The IPCE spectra were taken for the two DSSC systems, (A) $FTO/SnO_2/dye/electrolyte/Pt$ and (B) FTO/AuNP@ $SnO_2/dye/electrolyte/Pt$. Light from a Bentham PVE300 system connected to a TMC 300 IPCE set up was used with a xenon arc lamp (150 W) in the wavelength range of 300–800 nm.

RESULTS AND DISCUSSION

UV-Visible absorption of Au NP and Indoline dye

The colloidal solution of Au NPs used in this study shows a reddish-pink color (Figure 1). The UV-visible absorption spectra of the indoline dye solution and the Au NPs colloidal suspension are depicted in Figure 2. As can be seen from this figure, Au colloid shows a surface plasmon resonance absorption band with maximum around 547.7 nm. From the position of this absorption maximum, the diameters of Au NPs were estimated to be in the 70–80 nm range (Haiss *et a.l*, 2007; Njoki *et al.*, 2007). It was confirmed by the particle size analyzing measurement that higher degree of size distribution (diameter) of Au NP was at 73 nm. With the change in the size and the shape



Figure 2: Optical absorption spectra of the sensitizing dye, Indoline D149, (Red colour, solid line) and colloidal Au NPs (Purple colour, dash-dotted line).

of nanoparticles, the colour of the colloidal solution also changes. The synthesized nanoparticle size depends on the reaction rate, the concentration of precursor solution, the temperature and pH of the reacting medium as well as on the mixing rate of the reactors. In this research, trisodium citrate solution was added rapidly into the boiling Au³⁺ solution, although it has been reported that small sized Au NPs can be prepared by dropwise addition of citrate solution (Dissanayake et al, 2016). The colour of the Au NP colloidal solution shown in Figure 1 is due to the surface plasmon (SP) resonance oscillations of free electrons in Au NPs. As seen in Figure 2, the absorption band maximum of the dye and the SP absorption band maximum of Au NP are at 530 nm and 548 nm, respectively, and the Au NP absorption is much broader. Since, the SP oscillation frequency of Au is in the visible region, it enhances the light capturing in the wavelength range which matches well with the light absorption region of the Indoline dye (Hägglund et al., 2008), eventually increasing the photocurrent density which has been observed with the current - voltage characteristics of the solar cells.

J–V characterization and optimization of the amount of gold nanoparticles

Figure 3(b) shows the photocurrent density (J_{sc}) of the solar cells fabricated with photoanodes with different amounts of Au together with the pristine SnO₂ photoanode. Corresponding J_{sc} values and efficiency values plotted against the amount of Au nanoparticles incorporated are shown in Figure 3(a). It can be seen from the figure that both the J_{sc} and the efficiency show the maximum, corresponding to the drop–casted volume of 2.0 µL Au NP colloidal solution incorporated into the SnO₂ photoanode. The corresponding J-V characteristics curve is shown in Figure 3(b) in red in colour with half–filled–circles. Both the photocurrent density and the efficiency have first

increased with increasing amount of Au nanoparticles added to SnO₂ electrode, and then decreased with the further addition of Au nanoparticles. The enhancement in $J_{\rm sc}$ and the efficiency observed initially is very likely due to the LSPR effect leading to enhanced light absorption by the plasmonic Au NPs which is related to the number of Au NPs in the SnO₂ photoanode. However, with further increase in Au NP beyond the optimum value, both J_{SC} and efficiency decrease with the drop-casted amount of Au NP. This is very likely be due to the increase in parasitic absorption which does not generate photo-electrons and also due to the decrease in available sites for the dye adsorption caused by the presence of extra amount of Au NPs (Chao et al., 2015; Dissanayake et al., 2016; Chandrasekharan et al., 2000). The values of $J_{\rm SC}$, $V_{\rm OC}$, *FF* and efficiency η are shown in Table 1. By comparing the performance of the two DSSCs fabricated with two different photoanodes (bare SnO₂ and 2 µL AuNP@SnO₂) given in Table 1, it is clear that the plasmonic effect due to Au NPs have increased J_{sc} by an impressive 41.8%. In previous sections the plasmonic effect by Au NPs on EIS spectra and IPCE spectra has been reported and found that the addition of Au NPs to SnO, photo-electrodes has increased the photoconductivity, and reduced the charge recombination in the photoanodes, and improved the number of photogenerated electrons inside the solar cells, thereby increasing the of efficiency (η) of DSSCs by 26.8%.

EIS Characterization

Figure 4(a) shows the Electrochemical Impedance Spectra (EIS) of the DSSCs fabricated with bare SnO_2 (x=0) and AuNP@SnO₂ photoanodes. As already discussed (Dissanayake *et al.*, 2021), each Nyquist plot in Figure 4(a) shows the series resistance (R_s) and two semicircles (R_{CT1} and R_{CT2}). R_s is the external series resistor which occurs in the high frequency region. The larger semicircle at low

frequencies and the smaller semicircle at high frequencies of each fitted circuit consists charge transfer resistance $(R_{\rm CT2})$ at the TiO₂ photo anode/redox electrolyte interface, the back electron transfer resistance $(R_{\rm CT1})$ interface of redox electrolyte and Pt counter electrode respectively. *CPE*₁ and *CPE*₂ are constant phase elements of the capacitance corresponding to $R_{\rm CT1}$ and $R_{\rm CT2}$, respectively. The values of $R_{\rm s}$, $R_{\rm CT1}$ and $R_{\rm CT2}$ calculated from each fitted circuit are shown in Table 2. According to the table, the values of $R_{\rm s}$ and $R_{\rm CT2}$ of plasmonic DSSC made with AuNP@SnO₂ are lower than those of DSSCs made up of bare SnO₂. The resistance $R_{\rm s}$ is interpreted as the overall series resistance

of the device and the effects associated with deformations in the photoanode material. Further, it is also influenced by the inter-particle connectivity of the photoanode (Roy *et al*, 2010; Elumalai *et al.*, 2012). As shown in Table 2, R_s is lower for the plasmonic solar cell. This could be related to the enhancement of photovoltaic performance due to the enhanced photoconductivity by the metallic property of gold nanoparticles. Also, R_{CT2} has a value of 45.4 Ω for bare SnO₂ based solar cells and 33.7 Ω for the AuNP@SnO₂ based plasmonic solar cell, indicating that the presence of the noble metal nanoparticles, The presence of Au NPs in the photoanode has improved the electron transport process



Figure 3: (a) Photocurrent density and efficiency variation of the DSSCs with respect to the amount of Au NP colloidal solution added. (b) *J–V* characteristics for the series of DSSCs. used to optimize the PV parameters.

Table 1: I-V characteristics of plasmonic DSSCs made with Au NP incorporated SnO₂ photoanodes as a function of added amounts of gold nanoparticles.

SnO ₂ +x (µL) AuNP	$V_{\rm oc}({\rm mV})$	$J_{\rm SC}$ (mA cm ⁻²)	FF (%)	Eff (%) 2.28	
0	543.5	6.48	64.6		
1	544.4	8.23	60.4	2.71	
2	548.7	9.19	57.4	2.89	
3	550.9	7.34	59.5	2.41	
4	528.4	6.82	60.6	2.18	



Figure 4: (a) Nyquistic plots and (b) Bode phase plots for optimized DSSCs made with bare SnO, and AuNP@SnO, photoanodes.

 Table 2: Interfacial photoelctrochemical resistances and electron life time from EIS measurements of DSSCs made from two different SnO₂-based photoanodes.

Solar Cell	$J_{ m sc}$	V _{oc}	R _s	R _{CT1}	R _{CT2}	$f_{\rm max}$	electron life time $(\tau_e) / ms$
DSSC with Bare SnO ₂	6.48	543.5	19.0	6.56	45.4	18.57	8.57
DSSC with AuNP@ SnO ₂	9.19	548.7	12.0	11.20	33.7	23.90	6.66

by reducing the charge-transfer resistance R_{CT2} (by 35%) across the AuNP@SnO₂/electrolyte interface compared to the bare-SnO₂/electrolyte interface. According to Hägglund et al, this also will contribute to improve the electronic transport in the plasmonic solar cell, and this is the possible origin of the observed LSPR enhanced photoconductivity for DSSCs, which need to be considered as candidates for the plasmonic contribution to the short circuit current (Hägglund et al., 2008; Dissanayake et al., 2016). The $J_{\rm SC}$ of AuNP@SnO₂ based DSSC is higher, and the $R_{\rm CT2}$ value is lower compared to non-plasmonic DSSCs. This is evidently due to the enhanced photo-electron generation, better inter grain connectivity and creation of more conducting pathways in the SnO₂ photoanode by the metal AuNPs. This clearly indicates that the electron transport process is improved by the incorporation of AuNPs in to the SnO₂ photoanode. Figure 4(b) shows the corresponding bode phase plots extracted from EIS spectra. The value of the effective electron lifetime (can be estimated from the maximum frequency corresponding to the two DSSCs made with two different photoanodes, namely the bare SnO₂ and AuNP@SnO₂ using the following equation (Bisquert, 2002).

$$\tau_e = \frac{1}{\omega_{max}} = \frac{1}{2\pi f_{max}} \tag{1}$$

The corresponding values of $J_{\rm SC}$, $V_{\rm OC}$, and calculated of the DSSCs with bare–SnO₂ and AuNP@SnO₂ photoanodes are shown in Table 2. The values of $f_{\rm max}$ are 18.57 Hz and

23.90 Hz for the DSSCs made with bare SnO₂ and with AuNP@SnO₂ respectively. From Table 2, it is clear that the effective electron life time is lower in the plasmonic DSSCs. As the impedance measurements were taken under the exposed light condition, shorter life time means easy charge-transfer, faster photoconductivity and lower recombination. Therefore, the Au@SnO₂ photoanode has led to the enhancement in $J_{\rm SC}$, $V_{\rm oc}$ and the efficiency of the plasmonic solar cells. Similar results and explanation have already been reported (Gangadharan *et al.*, 2016).

IPCE measurements

The effectiveness of a DSSC cell to convert light of various wavelengths into electrical energy is measured by the incident photon-to-current conversion efficiency (IPCE), which is defined as the number of electrons generated by light per number of photons incident on the cell. The IPCE measures how efficiently the solar cell converts the incident photons into current at a given wavelength. Figure 5 shows IPCE curves of DSSCs with photoanodes made from bare SnO₂ film and AuNP incorporated SnO₂ film under the same illumination conditions. The higher IPCE values were observed for AuNP incorporated SnO, cell for almost over the entire wavelength range. An obvious feature of IPCE is the spectrum broadening to a longer wavelength upon AuNP incorporation on SnO₂, which is attributable to the plasma resonance effect (Li et al., 2013). IPCE expressed by equation (2) is defined as the number of photo-electrons generated by incident light per number of



Figure 5: Comparison of spectral responses (IPCE) of DSSCs made with two different photoanodes, bare SnO₂ and plasmonic AuNP incorporated SnO₂

photons. Likewise, IPCE spectrum can be used to calculate J_{sc} . This is done using the equation (3) and integrating the IPCE(λ) over the wavelength (λ) of the incident light (Xia–Zhio *et al.*, 2012),

$$IPCE\% = \frac{1240 \times J_{SC}}{\lambda \times W_i}$$
(2)

$$J_{SC} = \int q \times F(\lambda) \times IPCE(\lambda) \times d\lambda \tag{3}$$

where $J_{\rm SC}$ is the short-circuit photocurrent density (mA cm⁻²), λ the excitation wavelength (nm), Wi is the photon flux (W m⁻²), q is the electronic charge, and $F(\lambda)$ is the incident photon flux density at wavelength λ (Xia–Zhio *et al*, 2012; Li *et al*, 2013). The values of integrated $J_{\rm SC}$ for two different DSSCs made with bare–SnO₂ and AuNP@ SnO₂ photoanodes are tabulated in the inset of the Figure 5. According to this figure, both calculated $J_{\rm SC}$ (inset table of Figure 5) and directly measured $J_{\rm SC}$ (Table 2) values are consistent.

CONCLUSION

In this study, the effect localized surface plasmon resonance effect due to incorporation of Au nanaoparticles in SnO₂ photoanodes on the efficiency enhancement of the DSSCs fabricated with SnO₂ photo anode, sensitized with indoline dye was investigated. The addition of gold nanoparticles increased $J_{\rm SC}$, and hence, the overall conversion efficiency of the DSSC. It was found that the optimized amount of Au nanoparticles incorporated enhances the efficiency by more than 26%. Further incorporation of plasmonic AuNPs reduces the charge transfer resistant at SnO₂/electrolyte interface by 25.7% and also reduces the effective electron life time from 8.5 ms to 6.66 ms, thereby improving the charge transfer rate and electron-transport process. Thus, plasmonic AuNP incorporated SnO₂ based photoanodes can produce the DSSCs with significantly higher power conversion efficiency.

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DECLARATION OF CONFLICT OF INTERESTS

The authors declare no conflict of interests.

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