CHAPTER VI

EFFICIENCY STUDIES OF SOLANUM PROCUMBENS (SP), SOLANUM TORVUM (ST), ARTABOTRYS HEXAPETALUS (AH), GALINSOGA PARVIFLORA (GP) AND JASMINUM GRANDIFLORUM L (JG) PIGMENTS AS A SENSITIZER IN PT FREE GRAPHENE OXIDE, GRAPHENE OXIDE / NICKEL OXIDE AS COUNTER ELECTRODE FOR DYE SENSITIZED SOLAR CELL APPLICATIONS

6.1 INTRODUCTION

The ever-increasing need for energy necessitates swift action to efficiently utilize renewable energy sources. A dye-sensitized solar cell, also known as a thin film solar cell, is a low-cost solar cell. Even in low-light situations like indirect sunshine and cloudy sky also it works. The dye sensitised solar cell's operating principle is based on the photo production of an electron by a dye, similar to photosynthesis, and the dyes play an important part in enhancing the solar cell's efficiency. The (natural) dye sensitised solar cell is an attempt to replicate nature's photosynthesis process among many solar cells improving the power efficiency that lowers the cost of devices as compared to traditional energy sources. Due to its low cost, flexibility, relatively high efficiency, ease of production, and low toxicity, dye sensitised solar cells (DSSCs) have received a lot of attention in the recent two decades. According to recent DSSC advancements, the photo conversion efficiency is 14 %, and it still needs to be improved to reach the Schockley-Queisser limit of 33.7 % [1].

Graphene has a large surface area, outstanding electron transport properties, high mechanical strength, and high thermal and electrical conductivities. Because of its unique physico-chemical properties, it has a lot of potential applications for developing new approaches and making significant improvements in the field of electrochemistry. Graphene oxide/ metal oxide nanocomposites have aroused wide interest as hybridization improves the catalytic performance of graphene materials. Graphene oxide sheets help to anchor semiconducting particles and improve the performance of optoelectronic and energy conversion devices. Currently Nickel oxide nanoparticles are emerging as an alternative electrode on DSSCs because of their facile and reliable synthesis and characterization properties. They are most desirable in designing ceramic, magnetic, electro chromic and several electron transport applications. Latest reports have revealed that NiO combined with graphene sheets strengthen its mechanical stability [2].

In recent studies eco-friendly and low cost natural dyes have been employed but still an expensive metal such as platinum have been used as a counter electrode due to its high catalytic activity. There is a need to explore a counter electrode that is less expensive and non-corrosive electrolyte. Hence an attempt is made to use graphene oxide/ Nickel oxide as a counter electrode. The sensitizer plays a vital role in the functioning of DSSCs and mainly defines its performance. The dyes with a broader absorption range are required for efficient light harvesting [3].

The synthetic dyes N-719, N-749, and N-3 are used in the high-efficiency DSSCs that are expensive. Hence natural dyes, a widely available plant extract has been chosen as sensitizers for DSSCs such as Solanum Procumbens (SP), Solanum Torvum (ST), Artabotrys Hexapetalus (AH), Galinsoga Parviflora (GP) and Jasminum Grandiflorum L (JG). Solanum Procumbens (SP) extract have compounds like polyphenols, terpenoids, slenoids, and alkaloids, Solanum Torvum (ST) contains compounds like steroidal gluco-alkaloid and solasoine, Artabotrys Hexapetalus (AH) extract contains alkaloids, anthocyanins, and flavonoids, Galinsoga Parviflora (GP) extract contains electron-rich polyphenols, amino acids, anthocyanins, as well as sugars and iron content. Jasminum Grandiflorum L (JG). extract contains compounds like chlorophyll, betanins, anthocyanins, and carotenoids. Hence in this chapter, natural dye extracts from SP, ST, AH, GP and JG are used to prepare sensitizers for dye-sensitised solar cell fabrications.

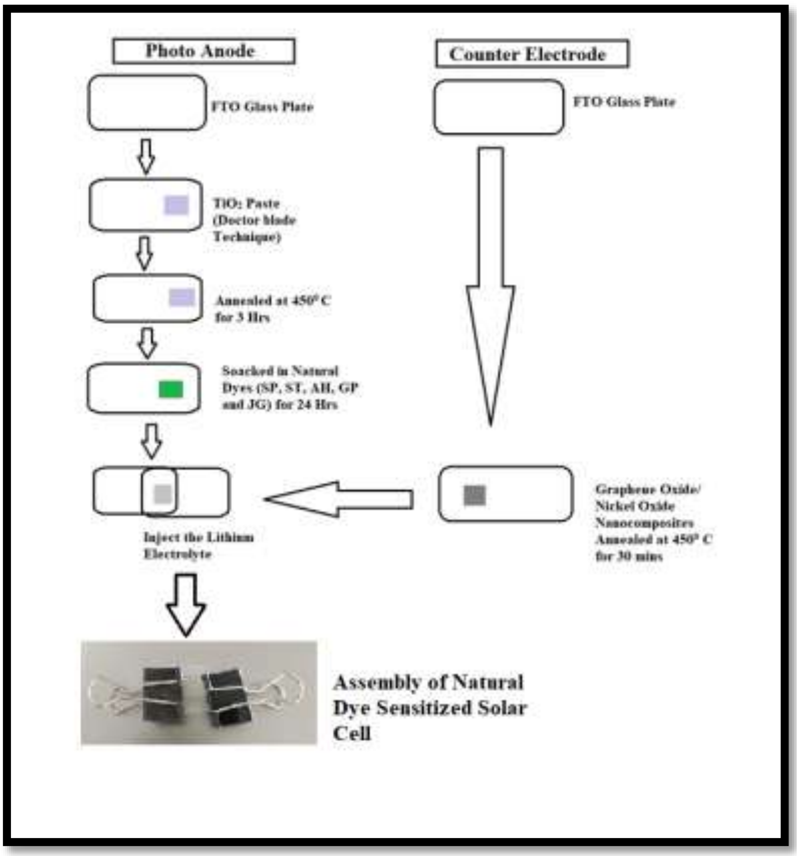
6.2. MATERIAL AND EXPERIMENTS

6.2.1 Natural Dye preparation

Fresh Solanum procumbens (SP) leaves are used for the extraction of constituents.10 grams of Solanum procumbens (SP) are mixed with 10 ml of distilled water and 10 ml of ethanol, kept under 45°C for 1 hour and left undisturbed for 24 hours at room temperature. The extract is filtered and used as a sensitizer in dye sensitized solar cell. Similar procedure is followed for Solanum Torvum (ST), Artabotrys Hexapetalus (AH), Galinsoga Parviflora (GP) and Jasminum Grandiflorum L (JG) leaves for the preparation of natural dyes.

6.2.2 Preparation of DSSCs

In an ultrasonic cleaner, the fluorine doped Tin oxide glass substrates are cleaned in distilled water, ethanol, and acetone for around 10 min each. To make a sticky past, mix 0.5 TiO_2, 3 mL ethanol, 1 g acetic acid, 10 CP ethyl cellulose, and 1.4 g Terpineol at 60° C for 48 hours. TiO_2 paste is applied with a doctor blade to a cleaned FTO glass plate and annealed at 450^o C. The active area of the electrode is 0.2cm. The dye extract (AH) is soaked in the prepared photo anode for 24 hours. The prepared nanocomposites (GO/NiO) of various concentrations (5:1, 5:2, 5:3, 5:4 and 5:5) are coated by using doctor blade method on FTO glass plate and annealed at 150^oC, acting as a CE. The ionic electrolytes (lithium iodine, iodine, acetonitrile and 2 dimethyl – 3Propylimidazolum iodine are injected between the sandwiches and are allowed to dry at room temperature [4].



6.1 Schematic Flow chart for the fabrication of Natural Dye Sensitized Solar cell using GO/NiO nanocomposites as counter electrode

6.2.3 Characterisation Techniques

The UV Vis absorption study is performed at room temperature using UV-Vis spectrometer (Cary 60 UV-Vis). The photo current studies are carried out in SP 150 biologic instrument with help of simulated solar light source.

6.3. RESULTS AND DISCUSSION

6.3.1 UV-Vis Spectral Analysis

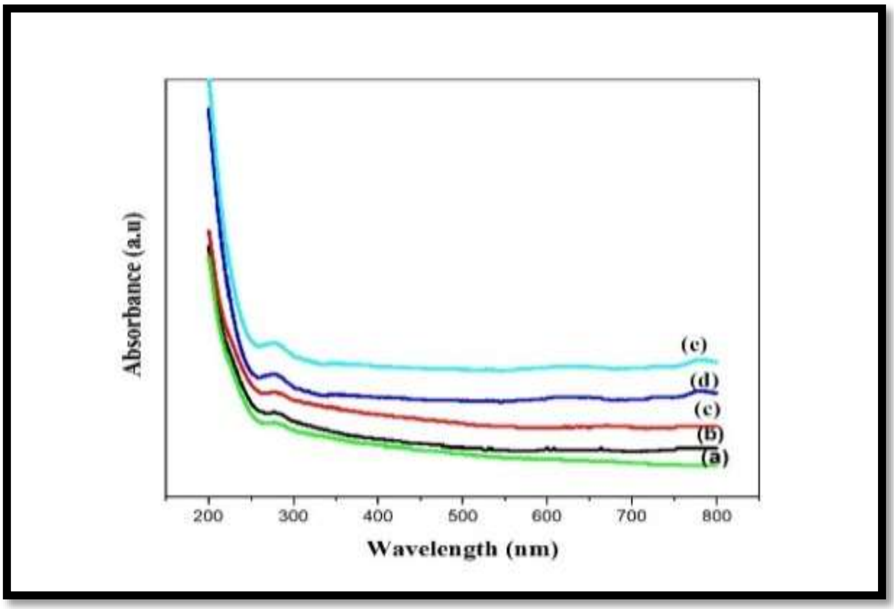


Figure 6.2 UV-Vis spectral analysis of GO / NiO of various concentrations (a) 5:1 (b)5:2 (c) 5:3 (d) 5:4 and (e) 5:5 nanocomposites

The optical properties of the GO/NiO of various concentrations (5:1, 5:2, 5:3, 5:4 and 5:5) nanocomposites are examined by UV-Vis Spectrophotometer within the range of 200 nm-600nm and are as shows in the Figure 6.2 (a-e). It shows that the absorption peak of GO/NiO for various NiO concentration are (5:1, 5:2, 5:3, 5:4 and 5:5) at 271 nm and 305 nm, 269 nm and 298 nm, 271 nm and 301 nm, 284 nm and 316 nm, 297 nm and 338 nm respectively. It is observed that the peaks are shifted to higher wavelength. This red shift may be due to the higher concentration of NiO on graphene nanosheets as evident from EDX and also the increase in the crystallite size as the concentration of NiO increases as evident from XRD analysis as discussed in Chapter III, as the movement of the electron in the nanoparticles, its size and shape plays a vital role in shifting the absorption peaks [5].

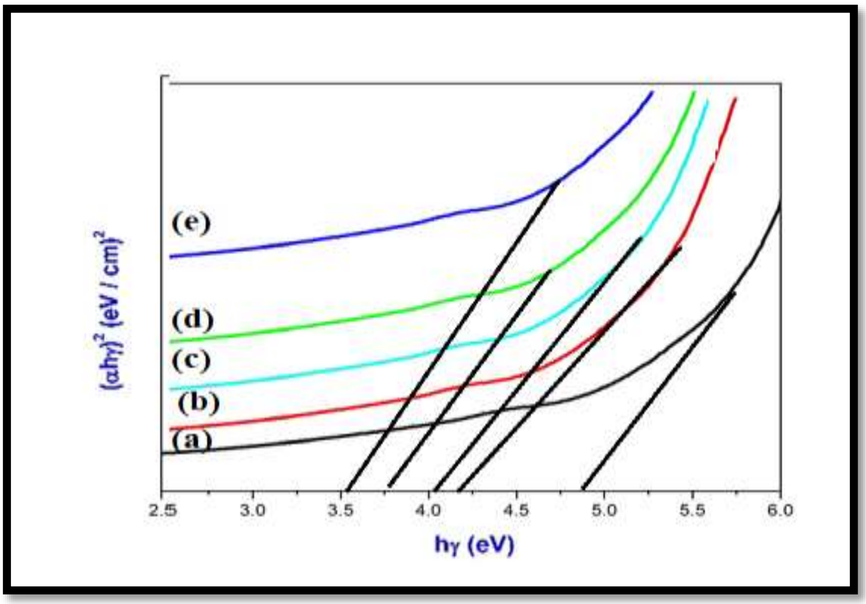


Figure 6.3 Band gap energy of Nanocomposites GO/NiO of various concentrations of (a)5:1, (b)5:2, (c) 5:3, (d) 5:4 and (e) 5:5

The band gap for GO/NiO (5:1, 5:2, 5:3, 5:4 and 5:5) nanocomposites is calculated from the Tauc plot and is shown in the Figure 6.3. The band gap energy is determined by using the equation.

$$\alpha = \frac{\alpha_0 (h\gamma - Eg)^n}{h\gamma}$$

where α_0 is a constant, Eg is the optical band gap and n is a constant, which depends on the probability of transition (i.e) n=1/2 for direct band gap [6,7].

The band gap energies for GO/NiO (5:1, 5:2, 5:3, 5:4 and 5:5) nanocomposites are found to,3.6 eV, 3.8 eV, 4.0 eV, 4.3 eV and 4.7 eV respectively. It is observed from the Figure 6.3 that the band gap increases due to the presence of NiO nanoparticles concentration onto the surface of Graphene oxide nanosheets. This red shift attributes to the increase in crystallite size of the GO / NiO (5:1, 5:2, 5:3, 5:4 and 5:5) nanoparticles and could also be evidenced from XRD analysis and the size of the nanocomposites has a great influence on the band gap [8].

6.3.2 Electrochemical Impedance Spectroscopy Analysis

The EIS Characteristics of the Dye Sensitized Solar Cell are investigated in this study. The EIS Nyquist plot of Pt free GO/NiO (5:1, 5:2, 5:3, 5:4 and 5:5) nanocomposites counter electrode with SP, ST, AH, GP and JG Dyes as a sensitizer for DSSCs device are shown in the Figures 6.4 (a-e). EIS is useful in analysing charge transport characterization of various interfaces in DSSCs devices, such as electron transfer and charge recombination into the TiO_2 / Dye / electrolyte interface, electron transfer impedance in the TiO_2 electrode, charge recombination in the counter electrode / electrolyte interface, and I_3^- / I^- transfer in the electrolyte [9]. Figure 6.4 (a-e) shows that the frequency range studied is divided into three parts. When light is applied to the DSSCs, the small circle in the lowest frequency corresponds to an ion diffusion impedance in the electrolyte (W_D), the large semicircle in the intermediate frequency corresponds to the charge transfer impedance in the TiO_2/ dye/ electrolyte interface (R_{tc}), and the smaller semicircle in the highest frequency corresponds to the counter electrode/ electrolyte interface (R_{tc}), and the smaller semicircle in the highest frequency corresponds to the charge transfer in the Counter electrode/ electrolyte interface (R_{CE}). The EIS results revealed that the GO/NiO (5:4) with JG Dye sensitizer achieves the highest semicircle in the highest frequency region. In the intermediate frequency region, the counter electrode had the lowest impedance value in the semicircle lesser than the GO/NiO (5:1. 5:2, 5:3 and 5:5) with SP, ST, AH and GP this is due to the higher NiO complexes in the GO/NiO (5:4), which have more bonding sites with the GO surface. The counter electrode made of GO/NiO (5:4) composites could allow for rapid electron transfer between the GO and NiO nanoparticles. The reduction of I^-_3 into I^- caused electrons to diffuse to the electrolyte, resulting in the NiO metal nanoparticles that can accelerate the diffusion of the I^-/I^-_3 redox pair, thereby enhancing the electro catalytic performance. This results showed that the increasing order of electron life time of the extracted dyes are SP <ST< AH< GP< JG. The longer electron lifetime is observed for the DSSCs using JG nature dye as a sensitizer when compared to the extract sensitizers SP, ST, AH and GP based DSSCs. It indicates that JG based DSSCs are more effective suppression of injected electrons recombination with I^-_3 in the electrolyte and that may be the cause for enhancement in the counter electrode and the device efficiency [10].

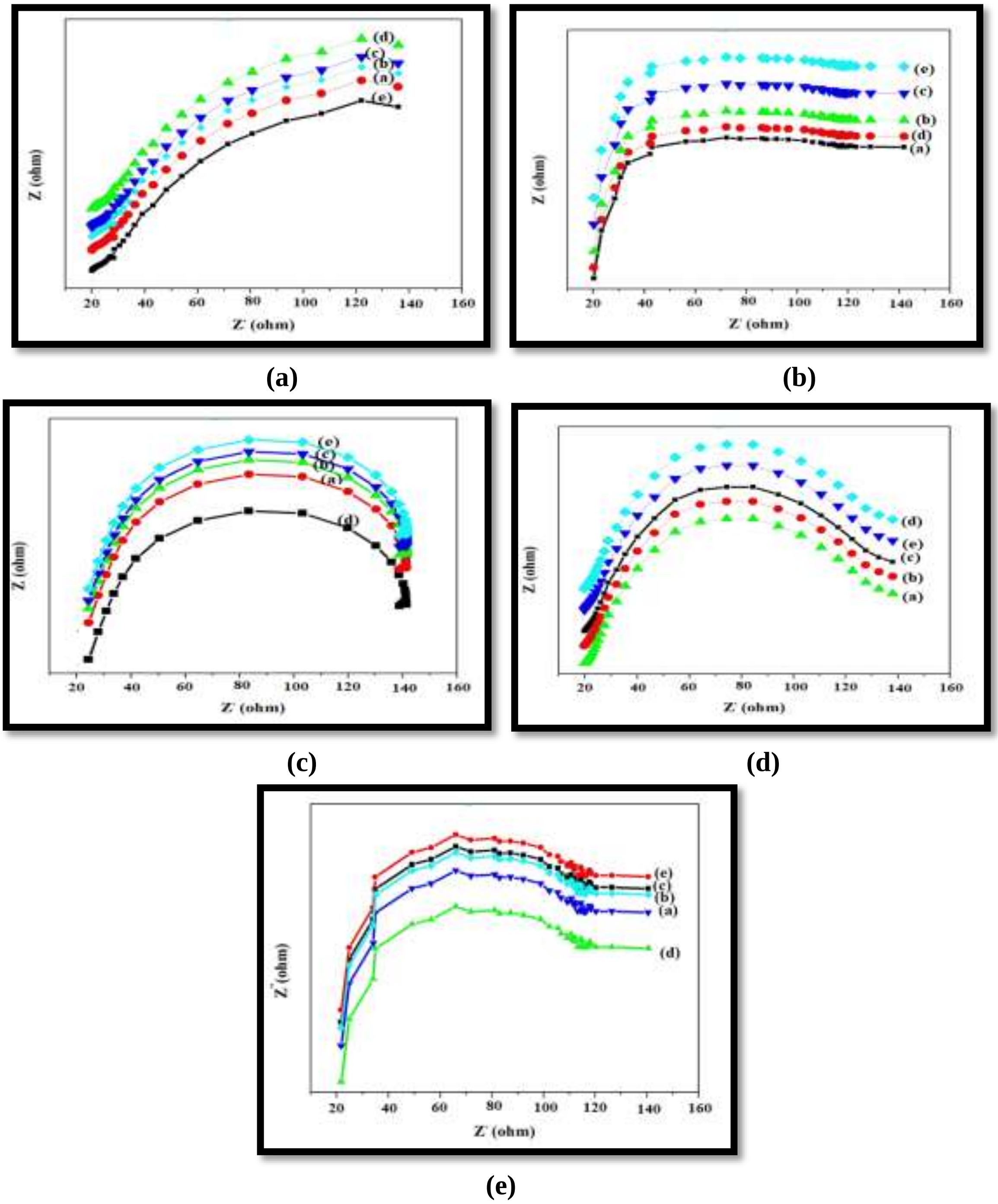


Figure 6.4 (a-e) Electrochemical impedance spectroscopy of a (a)GO/NiO (5:1, 5:2, 5:3, 5:4 and 5:5) counter electrode with SP as a sensitizer (b) GO/NiO (5:1, 5:2, 5:3, 5:4 and 5:5) counter electrode with ST as a sensitizer (c) GO/NiO (5:1, 5:2, 5:3, 5:4 and 5:5) counter electrode with AH as a sensitizer (d)GO/NiO (5:1, 5:2, 5:3, 5:4 and 5:5) counter electrode with GP as a sensitizer (e) GO/NiO (5:1, 5:2, 5:3, 5:4 and 5:5) counter electrode with JG as a sensitizer

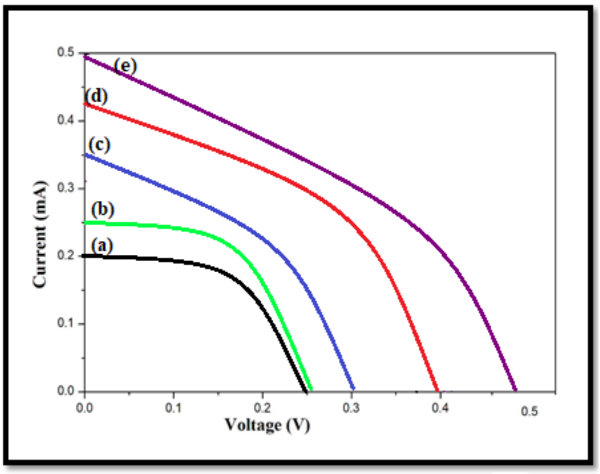
6.3.3. Efficiency Studies

Photovoltaic characterization of the prepared nanocomposites is investigated by I-V studies. The prepared nanocomposites are used as a counter electrode and natural extract is used as a dye sensitizer. The effective area of the DSSC are measured to be 0.2 cm and the performance of DSSC are evaluated by short circuit current density(I_{sc}), open circuit current (v_{oc}) fill factor (FF) and the energy conversion efficiency (η) are calculated by using the equations (1) and (2).

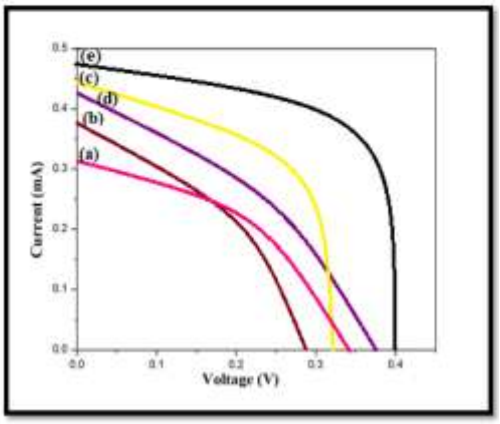
$$FF=(I_{max} x V_{max}) / (I_{sc} x V_{oc}) \quad (1)$$

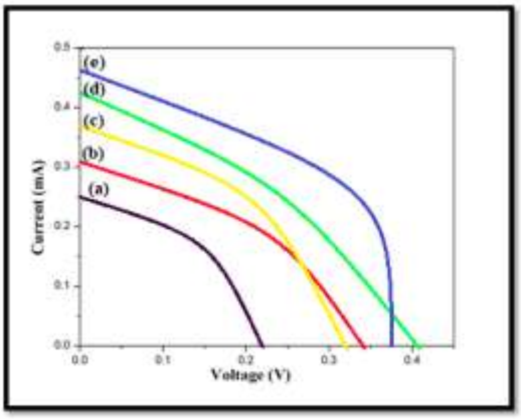
$$\eta = J_{sc} V_{oc} FF / P_{in} x 100 \quad (2)$$

where I_{max} and V_{max} denotes the maximum output value of current and voltage respectively and I_{sc} is the short circuit current density. The performance of SP, ST, AH, GP and JG extract as dye sensitizer are evaluated by determining the open circuit current, short circuit current, Fill Factor and conversion efficiency under the irradiance of 100 W/cm^2. The typical I-V curves of Dye sensitized solar cell is shown in the Figure 6.5 (a-f) and. Figure 6.5 (a-f) shows that the I-V characteristic spectrum performance of GO and GO/NiO (5:1 5:2, 5:3, 5:4 and 5:5)as a counter electrode with SP, ST, AH, GP and JG extract as a dye sensitizer [11].

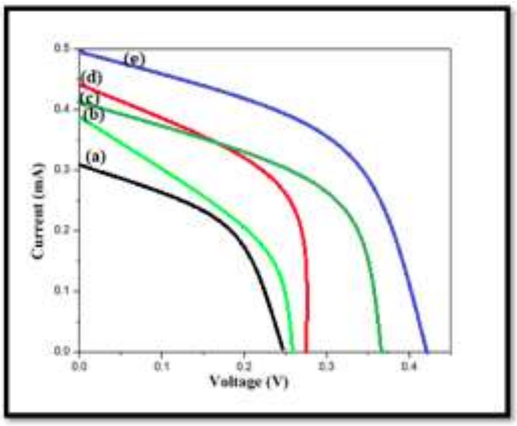


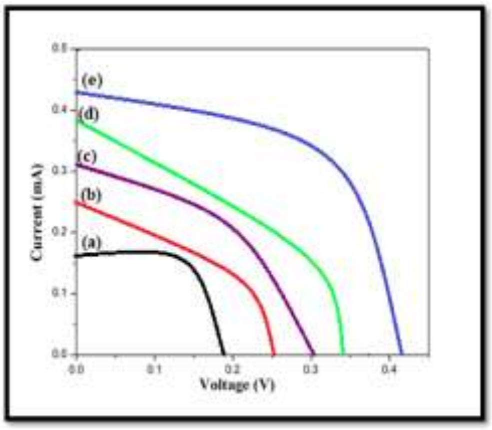
(a)



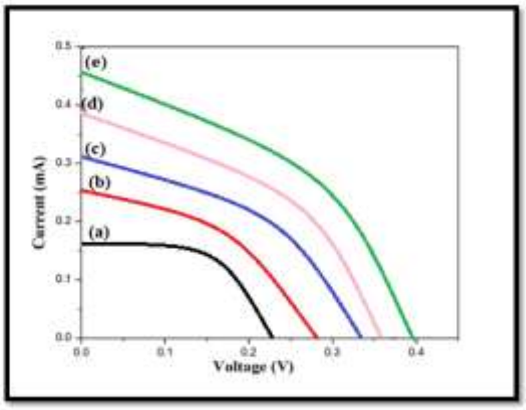


(b) **(c)**





(d) **(e)**



(f)

Figure 6.5 Current density Voltage (I-V) curves of (a) GO with SP, ST, AH, GP and JG dye (b-f) GO/NiO (5:1, 5:2, 5:3, 5:4 and 5:5) with SP, ST, AH, GP and JG Dye

Figure 6.5 (a-f) shows that DSSC I-V curves of the prepared GO and GO / NiO (5:1, 5:2, 5:3, 5:4 and 5:5) nanocomposites counter electrode with SP, ST, AH, GP and JG dyes as a sensitizer for platinum free DSSCs. Figure 6.5 (a) shows that Graphene oxide as a counter electrode with JG dye extracts as a sensitizer achieve better efficiency than the other dyes and it may be due to JG comprises of many compounds, like terpenoids, alkaloids, flavonoids and anthocyanin which is the foremost mixtures of high chlorophyll pigment which occurs in the light reaction of photosynthesis. Figure 6.5 (b-f) shows that GO/NiO (5:4) Counter electrode with JG sensitizer achieves better efficiency. The efficiency of natural dye based DSSCs is correlated with absorption of the dye and anchoring of the dye molecule onto the anode (TiO_2) surface. Higher interaction between TiO_2 and dye molecule leads to better charge transfer. Figure 6.5 (f) also confirms that the GO/NiO (5:4) with JG sensitized CE archives the high electron transfer and low impedance DSSCs [12]. Hence the prepared GO/NiO (5:4) nanocomposites act as a counter electrode, that is corrosion free and with natural dye as a sensitizer achieves high efficiency of 2.08%.

Table 6.1 Photovoltaic parameters of the GO nanosheet as counter electrode with natural extracts of SP, ST, AH, GP and JG as a sensitizer for DSSC

Dye	**Counter Electrode**	**V_{oc}(mV)**	**I_{sc}(mA)**	**Fill Factor**	**Efficiency**
SP	GO	0.3	0.31	33%	0.3%
ST	GO	0.32	0.3	34%	0.7%
AH	GO	s0.33	0.36	36%	0.65%
GP	GO	0.35	0.33	38%	0.8%
JG	**GO**	**0.36**	**035**	**41.1%**	**0.96%**

Table 6.2 Photovoltaic parameters of the GO/NiO (5:1 to 5:5) nanocomposites counter electrode and SP as a sensitizer for DSSCs

Dye	Counter Electrode (concentration) GO/NiO	V_{oc}(mV)	I_{sc} (mA)	Fill Factor (%)	Efficiency (%)
SP	5:1	0.27	0.27	26%	0.42%
SP	5:2	0.3	0.3	27%	0.51%
SP	5:3	0.2	0.31	29%	0.57%
SP	5:4	0.36	0.4	31%	0.61%
SP	**5:5**	**0.3**	**0.3**	**33%**	**0.65%**

Table 6.3 Photovoltaic parameters of the GO/NiO (5:1 to 5:5) nanocomposites counter electrode and ST as a sensitizer for DSSCs

Dye	Counter Electrode (concentration) GO/NiO	V_{oc}(mV)	I_{sc} (mA)	Fill Factor (%)	Efficiency (%)
ST	5:1	0.2	0.26	29%	1.0%
ST	5:2	0.3	0.2	31%	1.2
ST	5:3	0.31	0.25	35%	1.23
ST	**5:4**	**0.3**	**0.3**	**39%**	**1.25**
ST	5:5	0.4	0.3	27 %	1.2

Table 6.4 Photovoltaic parameters of the GO/NiO (5:1 to 5:5) nanocomposites counter electrode and AH as a sensitizer for DSSCs

Dye	Counter Electrode (concentration) GO/NiO	V_{oc}(mV)	I_{sc} (mA)	Fill Factor (%)	Efficiency (%)
AH	5:1	0.25	0.3	30%	0.6%
AH	5:2	0.26	0.4	34%	0.65%
AH	5:3	0.3	0.4	35%	0.7%
AH	5:4	0.3	0.41	36%	0.89%
AH	**5:5**	**0.35**	**0.4**	**40%**	**0.9%**

Table 6.5 Photovoltaic parameters of the GO/NiO (5:1 to 5:5) nanocomposites counter electrode and GP as a sensitizer for DSSCs

Dye	Counter Electrode (concentration) GO/NiO	V_{oc}(mV)	I_{sc} (mA)	Fill Factor (%)	Efficiency (%)
GP	5:1	0.35	0.3	39%	1.81%
GP	5:2	0.36	0.36	41%	1.9%
GP	5:3	0.41	0.4	40%	1.95%
GP	**5:4**	**0.4**	**0.45**	**42%**	**1.97%**
GP	5:5	0.4	0.43	40%	1.8%

Table 6.6 Photovoltaic parameters of the GO/NiO (5:1 to 5:5) nanocomposites counter electrode and JG as a sensitizer for DSSCs

Dye	Counter Electrode (concentration) GO/NiO	V_{oc}(mV)	I_{sc} (mA)	Fill Factor (%)	Efficiency (%)
JG	5:1	0.4	0.4	36%	1.8%
JG	5:2	0.42	0.4	38%	1.98%
JG	5:3	0.43	0.41	40%	2.01%
JG	**5:4**	**0.45**	**0.4**	**43%**	**2.08%**
JG	5:5	0.4	0.36	42%	2.03%

Table (6.1-6.6) shows the efficiency of DSSC in the prepared Pt-free GO and GO/NiO (5:1, 5:2, 5:3, 5:4, and 5:5) nanocomposites as a counter electrode with SP, ST, AH, GP, and JG dyes as a sensitizer. Table 6.1 shows that the prepared Pt free GO nanosheet works as a counter electrode with SP, ST, AH, GP and JG dyes as a sensitizer for DSSC. It is observed from Table 6.1 that the prepared GO based counter electrode with SP, ST, AH, GP and JG dyes sensitizer achieves 0.3 %, 0.7 %, 065%, 0.8 % and 0.96%. efficiency respectively. The high conductivity material graphene oxide and large absorption dye sensitizer will enhance the DSSC efficiency. It is observed from the Table 6.2 that the GO/NiO (5:5) nanocomposites as a counter electrode with SP sensitizer achieve a maximum efficiency of 0.65%. Similarly GO/NiO (5:5) nanocomposites from Table 6.3 with ST sensitizer achieves the best efficiency of 1.25 %, and Table 6.4 denotes that the prepared GO/NiO (5:5) nanocomposites with AH achieve 0.9 % efficiency. Tables 6.5 and 6.6 show that GO/NiO (5:4) concentration nanocomposites achieves 1.97 % and 2.08 % efficiency respectively. Graphene oxide has low charge transfer resistance and excellent electro catalytic activity and these factors are responsible for higher current densities. In DSSC, the dyes absorb large amount of sun light to enhance the efficiency of DSSC, which is confirmed from UV-Vis analysis in chapter II as the JG dye archives the large absorption of 339 nm.

6.4 CONCLUSION

This chapter describes the preparation of Pt free GO and GO/NiO (5:1, 5:2, 5:3, 5:4 and 5:5) nanocomposites as a counter electrode and SP, ST, AH, GP and JG sensitized DSSC. The UV-Vis studies showed that the prepared GO/NiO (5:1, 5:2, 5:3, 5:4 and 5:5) nanocomposites absorption peak 271 nm and 305 nm, 269 nm and 298 nm, 271 nm and 301 nm, 284 nm and 316 nm, 297 nm and 338 nm respectively. The energy band gap of GO/NiO (5:1, 5:2, 5:3, 5:4 and 5:5) is 3.6 eV, 3.8 eV, 4.0 eV, 4.3 eV and 4.7 eV respectively. The EIS analysis discovered that the GO/NiO (5:4) nanocomposites with JG sensitized DSSC archives the low electron transfer resistance. The I-V characteristics showed that the photovoltaic efficiency of GO DSSCs with SP, ST, AH, GP and JG sensitizer are 0.3 %, 0.7 %, 0.65 %, 0.8 % and 0.96 % respectively and GO/NiO (5:1, 5:2, 5:3, 5:4 and 5:5) nanocomposites as counter electrode for DSSCs with SP, ST, AH, GP and JG as sensitizer achieves 0.6%, 1.25%, 0.9%, 1.97% and 2.08% respectively. GO/ NiO (5:4) counter electrode JG sensitizer efficiency is higher than SP, ST, AH and JG sensitizer in GO/NiO (5:1 to 5:5) based counter electrode DSSCs. This work confirms that the prepared GO/NiO (5:1, 5:2, 5:3, 5:4, and 5:5) nanocomposites as counter electrode has a low cost and environmentally friendly dyes (SP, ST, AH, GP, and JG) that make them attractive for their use as a sensitizer in dye sensitized solar cells.

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