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Remazol Orange Dye Sensitized Solar Cell using Graphene Oxide and reduced Graphene Oxide Working Electrode

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ABSTRACT

Dye Sensitized Solar Cell (DSSC) is said as a potential solar device which offers easy, cheaper and greener materials and preparation process. However, the efficiency of this device is still an ultimate problem and challenge. In this paper, an organic Remazol orange dye was used as the DSSC dye sensitizer which prepared with different working electrodes. The different working electrodes consist of Titanium Dioxide (TiO_2) with Graphene Oxide (GO) and TiO_2 with reduced Graphene Oxide (rGO). In order to analyze the adsorption characteristics of GO and rGO, the solution was tested using Ultraviolet-Visible-Near Infrared Spectrophotometry and the surface morphology of all mixed pastes was observed under Atomic Force Microscopy and Scanning Electron Microscope. Then, the device performance was tested under illumination of solar cell simulator. From overall results, the efficiency for all tested devices was quite low from expectation. For this work, the performance of TiO_2-rGO DSSC at 0.138% is 84.7% higher compared to the TiO_2-GO DSSC which was 0.021%. This result was obtained when the working electrode and dye less exposed to the light during dye preparation process at 24 hours soaking time.

Keywords: Graphene Oxide; Reduced-Graphene Oxide; DSSC; Remazol Orange Dye.

1. INTRODUCTION

Dye Sensitized Solar Cell (DSSC) is a photovoltaic device that incorporates organic and inorganic materials. This device is well known with its characteristics of low cost of manufacturing production, sensitization materials and green technology [1]. The sensitization of wide band material [2] and dye molecules [3] contribute to the conversion of the visible light to electrical energy. The dye sensitizer selection for the working electrode will show different intensity of absorption. The performance of solar cell is determined by the absorption of spectrum into dye and its attachment to the surface of Titanium Dioxide (TiO_2) that normally used as working electrode. The great binding energy between the dye sensitizer with the TiO_2 is required to produce excellent charge transfer adsorption in the visible range [4].

Ruthenium (Ru) sensitizer is the most established dye as it allowed charge transfer adsorption in wide adsorption range. But Ru sensitizer is very expensive and not environmental friendly [5]. As alternative to Ru, there are many natural dyes have been used such as Mangosteen pericarp [6] and beetroot [7] which have performed conversion efficiency up to 1.2%. Even though these natural dyes promised the low cost process but the result for the efficiency is low because the interaction between the pigment in the dye and TiO_2 would been low [8], easy to oxidized and difficult to control the concentration of the dyes.

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Although the work on DSSC has been performed in many years, the efficiency of DSSC is still a great challenge. There are many efforts that have been done in order to boost up the device performance including hybridizing all parts (die, working electrode, counter electrode) with conducting carbon based materials like carbon nanotubes [9], graphene [10]. carbon nanotubes-graphene [11] and even activated carbon like bamboo charcoal and activated bamboo charcoal [12]. In this study the Remazol Orange dye was used as the dye sensitizer based on work performed in [13]. In order to investigate the effect of different materials on the working electrode, the conventional working electrode which is TiO_2 will be mixed with graphene oxide (GO) and (rGO). The performances of these two devices will be analyzed and investigated.

2. MATERIALS AND METHOD

2.1 Dye Sensitizer

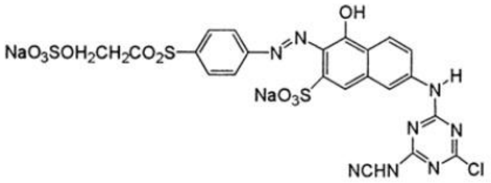


Figure 1. Chemical structure of Remazol Orange [13].

In this work, water based dye Remazol Orange was purchased from AR Alatan Sains Sdn. Bhd and used in this experiment. The chemical structure of Remazol Orange was shown in Figure 1. The concentration of 2.5 mM was used in this experiment by referring to [13].

2.2 Preparation of Working Electrode

An ITO glass of 2 x 2 cm^2 was prepared by cleaning it with ethanol (CH_3CH_2OH). The sheet resistance of ITO glass was 9 Ω/cm^2. The side with the resistance is faced up and covered with cellophane tape around 0.2 mm at the head and bottom of the glass and left only 1 x 2 cm^2 as an active area. The stabilizer was prepared by adding 0.1 mL of acetic acid ($C_2H_4O_2$) with 50 mL of DI water and stirred for 2 to 5 minutes. Then, it was left for few minutes to dissolve. A drop of stabilizer was then added to 1 g of Titanium dioxide (TiO_2) nanoparticles powder in a crucible lid in order to form a TiO_2 paste. 1 mL drop of Graphene Oxide (GO) or reduced Graphene Oxide (rGO) with the concentration of 9 mg/mL was dropped in the crucible lid followed by a drop of Triton X – 100. It was stirred slowly in order to make sure that the stabilizer and the TiO_2 mixed paste was fully blended. The crucible lid was covered for 5 minutes to make sure all materials were mixed well. The TiO_2 mixed paste was added onto the glass by using glass rod and distributed evenly on the glass surface by using Dr Blade deposition technique. The working electrode glass is baked for 450 °C for 30 minutes in vacuum furnace.

2.3 Fabrication of Dye Sensitized Solar Cell

The preparation of working electrode was continued by soaking either TiO_2-GO glass or TiO_2-rGO glass in the Remazol Orange dye solution. This process was done at three conditions; 24 hours uncovered (exposed to light), 2 hours uncovered and 24 hours covered. For the counter electrode, the ITO glass was held by facing it down on a light candle by using forceps. The soot from the combustion of flame of the candle resulted in a black deposition on the conductive glass. Then both of working and counter electrode were sandwiched by clipping together at the end of electrodes. A few drops of iodine electrolyte were dropped between the two electrodes.

2.4 Characterizations

The surface morphology of TiO_2-GO and TiO_2-rGO was observed under SPI3800N Atomic Force Microscopy (AFM) and Scanning Electron Microscope (SEM). The adsorption spectra of GO and rGO solution was characterized using Ultraviolet-visible-near infrared spectrophotometry (UV-Vis-NIR, Lambda 25,35,45, PerkinElmer). The performance of full set of DSSCs was examined under PV Cell Testing LS1000 Solar Simulator to illuminate the DSSC and a digital source meter (Keithley 2450) was used to obtain the Isc and Voc. The maximum voltage (open circuit voltage, V_{OC}), maximum current (short circuit current, I_{SC}) and fill factor (FF) were achieved from the experiment. The fill factor and overall efficiency were calculated using Equation 1 and Equation 2 respectively:

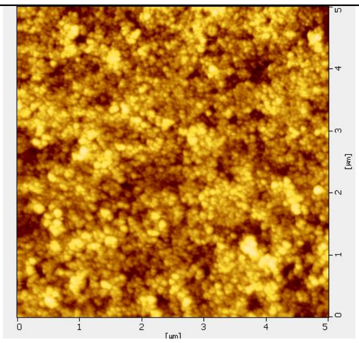
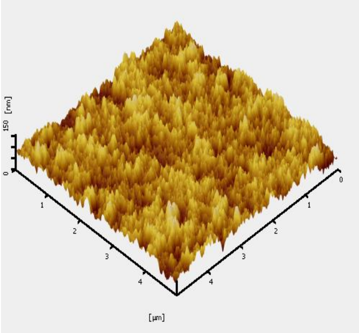
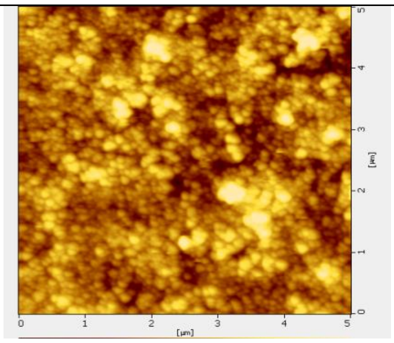
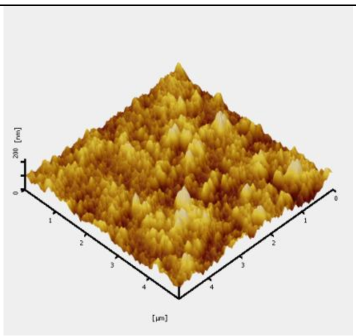
$$FF = \frac{P\max}{Isc \times Voc} \tag{1}$$

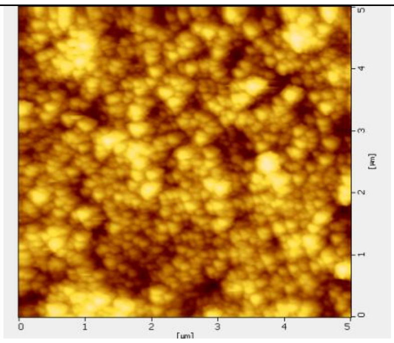
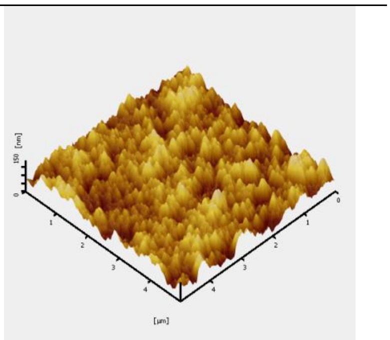
$$Efficiency, \eta = \frac{Jsc \times Voc \times FF}{Pin} 100\% \tag{2}$$

3. RESULT AND DISCUSSION

3.1 Atomic Force Microscopy

Table 1 AFM images of TiO_2, TiO_2 with GO and rGO

Sample	Surface area	3D Image
TiO_2		
TiO_2-GO		

TiO_2-rGO		

The surface morphology of TiO_2 with GO and rGO which observed using AFM was tabulated in Table 1. Based on the results, the surface of the three samples; TiO_2, TiO_2-GO and TiO_2-rGO were clearly observed. For TiO_2 sample, it showed that the average diameter of particle for the sample was 1 nm per particle and the shape of particle was even throughout the sample. The surface was a bit rough for the TiO_2 sample. For the TiO_2 -GO sample, the diameter of the particle is similar to TiO_2 with even and uniform shape. The overall surface was smooth but slightly rough at certain part. The diameter size of the particle for TiO_2- rGO samples was slight reduced to 0.5 nm per particle due to the less oxide element in rGO [14]. However, the particles were in even shape throughout the sample with smoother surface.

3.2 Scanning Electron Microscope

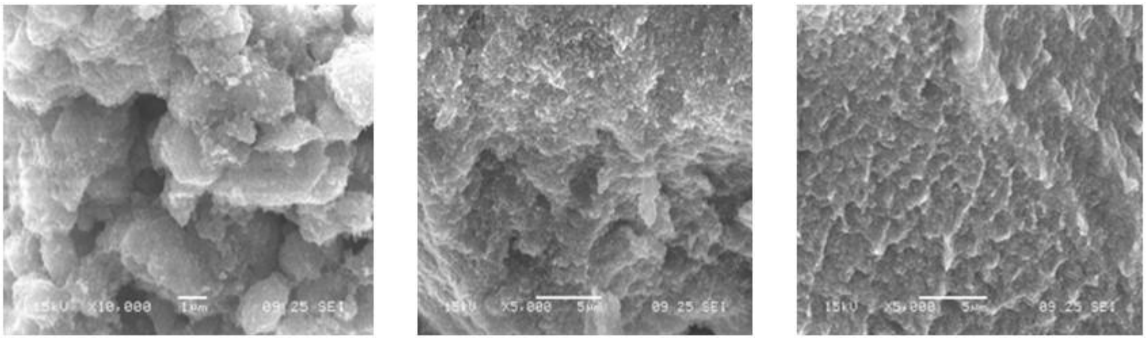


Figure 2. SEM images of (a) TiO_2, (b) TiO_2-GO and (c) TiO_2-rGO surfaces.

The surface morphology of TiO_2 with GO and rGO was next observed under SEM. Figure 2 (a) shows that the TiO_2 surface were rougher compared to TiO_2-GO and TiO_2-rGO as shown in Figure 2 (b) and (c) respectively. It was believed that the rougher surface of TiO_2 sample was resulted from the TiO_2 powder which was not well grinding during the preparation sample. The smoother surfaces were believed due to the addition of TiO_2 with GO or rGO which give the paste much lesser viscous rather than TiO_2. The smoothest particle was obtained for TiO_2 with rGO sample attributed to addition of the oxide from TiO_2 to the rGO which result a tightly bond structure [14].

3.3 Ultraviolet-Visible-Near Infrared Spectrophotometry

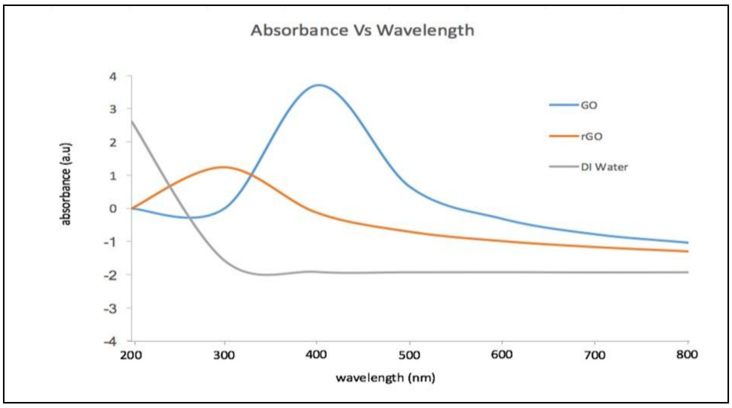


Figure 3. Uv-Vis graph of GO and rGO solution.

Ultraviolet-visible-near infrared spectrophotometry (UV-Vis) was used to identify absorption characteristic of the prepared solutions. The adsorption of Remazol Orange dye which observed at 560 nm was reported in early work [13] which comparable to the absorbance peaks of the tested organic dyes such as Roselle [15] at peaks of 320 nm and 520 nm, and 516 nm for red cabbage [16]. The adsorption of GO and rGO solutions were observed from the wavelength of 200 to 800 nm as shown in Figure 3. The result shows that the absorption peaks of 300 nm and 400 nm were recorded for rGO and GO respectively which attributed by the π-π* transition of the aromatic C-C bond [17].

3.4 DSSC Performance

Reported from the previous work [13] the efficiency of Remazol Orange DSSC at the concentration of 2.5 mM with TiO_2 working electrode was recorded at 1.07%. A similar result was obtained by using activated bamboo charcoal mixed in TiO_2 as the working electrode [12]. Table 2 summarized the efficiency of all DSSCs tested under three different dye preparation conditions; the working electrodes were soaking in the dye for 24 hours uncovered process (exposed to light), 2 hours uncovered and 24 hours covered from light.

Table 2. Efficiency of GO and rGO.

TiO_2-rGO	**TiO_2-GO**
24 hours uncovered (exposed to light)	
0%	0 %
2 hours uncovered (exposed to light)	
0.068 %	0.002 %
24 hours fully covered from light	
0.138 %	0.021 %

From Table 2, it was noticed that all efficiencies were quite low from the expectation, although the efficiency for TiO_2-GO DSSC and TiO_2-rGO DSSC was increased as the time of the dye solution exposed to light decreases. For TiO_2-GO and TiO_2-rGO working electrodes, both efficiencies are 0% as they were exposed to light for 24 hours. It was believed that this result was obtained due to the sensitivity of the dye to the light [18]. Once it was exposed to the light for a long duration, the quality of dye solution will be downgraded.

As expected, when the dye solution was exposed to the light for 2 hours (dye less exposed to the light), the efficiency of both DSSCs increased up to 0.068% and 0.002% for TiO_2-rGO and TiO_2-

GO respectively. It was assumed that once the TiO_2 was added to the rGO, it completes the rGO oxide group and make the connectivity to connect the electricity was much better than GO which makes the efficiency using TiO_2-rGO was greater than TiO_2-GO. In another hand, rGO was known as a conducting material compared to GO.

When the dye solution was fully covered from light for 24 hours before the device characterization, drastic changes have affected the efficiency of both DSSCs. The efficiency of DSSC was increased up 0.138% and 0.021% for TiO_2-rGO and TiO_2-GO respectively attributed to the covered dye solution process.

Although the results were improved by controlling exposure of the light, but still the efficiencies of these DSSCs were very low compared to the recorded results in [12,13]. There are some factors that may affect the device performance such as the mixture of TiO_2 with GO and rGO was unwell blended, low concentration of GO and rGO used during hybrid process, non uniform film during Dr Blade deposition technique, crack during baking and others. Further investigations need to be done on this matter as we expect better performance should be obtained for these mixtures compared to work reported in [12].

4. CONCLUSION

The performance of DSSC prepared at different working electrodes; TiO_2-GO and TiO_2-rGO was investigated. The surface morphology of both mixtures has been observed using AFM and SEM which shows the TiO_2 particles became more smoother with the mix of GO and rGO. The absorption peak at 300 nm and 400 nm are recorded for rGO and GO respectively. The best efficiency at 0.138% was obtained for TiO_2-rGO DSSC compared to TiO_2-GO DSSC which was recorded at only 0.021%. This device performance was improved when the working electrodes soaked for 24 hours in the dye and covered from the light before device characterization. However, the performance of these DSSCs were still low compared to the previous reported Remazol orange DSSC. Further enhancements are required in order to increase the DSSC performance.

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