The Application of Dye-Sensitized Solar Cell Using rGO and MBs in Series-Parallel Under Low Illumination

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ABSTRACT The advantage of the dye-sensitized solar cell (DSSC) is utilized indoor fluorescent light illumination, but the output power of DSSC is not high enough. In this study, we provided a structure for DSSCs, and characterized the photovoltaic performances under air mass 1.5 global and indoor fluorescent light illumination. The photoanode of DSSC is based on titanium dioxide (TiO2) – magnet beads (MBs) – reduced graphene oxide (rGO) composited photoanode (TMGP), which was fabricated by hydrothermal method, spin coating, and doctor blade. According to the experimental results, adding MBs and rGO to DSSCs can enhance the charge transfer ability, reducing the occurrence of charge recombination, thereby improving the photovoltaic performance. DSSCs with TMGP photoanode can maintain a photovoltaic conversion efficiency of around 14% under indoor light.

INDEX TERMS Dye-sensitized solar cells (DSSCs), electrochemical impedance spectroscopy (EIS), graphene, titanium dioxide (TiO2).

I. INTRODUCTION

The demand in energy supply has accelerated fossil fuel depletion. It is predicted that the reverses of fossil fuel can only last forty years, sixty years for natural gas, and two hundred for coal [1]. The renewable energy has been considered in recent years, such as photovoltaic, wind power, geothermal heat, hydropower, and biomass energy. Among all renewable energy technologies, the photovoltaic technology is the particularly promising technology for direct conversion sunlight into electricity energy [2], [3], [4], [5], [6]. The developments of the solar cells are able to be divided into four generations: (1) the silicon-based solar cells (single, polycrystalline, and amorphous silicon); (2) the thin-film cells (CdTe, CIGS, and CIS); (3) the organic matter and nanotechnology cells. However, silicon-based solar cells are high production costs and confine to application in terrestrial photovoltaic. In comparison with silicon-based solar cells and thin-film cells, the dye-sensitized solar cell (DSSC) has low manufacturing costs, high-temperature environment, transparency, flexibility, and generate electricity with indoor light sources [7], [8], [9], [10], [11]. Due to the lower illumination application, the DSSC can work efficiently in dark conditions, for example, dawn or dusk. Hence, DSSC can be the most promising photovoltaic device. The DSSC consists of three parts with the sensitized working layer (photoanode), electrolyte, and the counter electrode. In general, the photoanode material is titanium dioxide (TiO2), because the TiO2 has good compatible with dye molecules. Moreover, the photoanode of DSSC is acted as a very important role, which takes responsibility for transforming sunlight into electric power [12]. Therefore, the photovoltaic conversion efficiency
of DSSC is usually improved by modifying photoanode. Moreover, the topic of our research group is the enhancement of photovoltaic conversion which is ameliorated by modifying the nanostructure of active layer (semiconductor layer) or retardation of dark reaction at interface. For instance, Chou et al. in [13], the indium gallium zinc oxide (IGZO), the indium gallium zinc oxide (IGZO), IGZO barrier layer is based on the photoanode with DSSC via sputtering. It can retard dark reaction and back reaction to improve the photovoltaic conversion efficiency. Furthermore, Chou et al. in [14], graphene oxide (GO) and zinc oxide (ZnO) have been applied to photoanode to increase the amount of dye-loading and reduce electron transfer impedance, respectively. In addition, Chou et al. in [15], GO and magnetic beads (MBs) have also introduced into counter electrode for photocatalytic activity and fast electrocatalyst of triiodide reduction. Furthermore, Chou et al. in [16], the Fe3O4 has been applied to active layer for the decrease in electron recombination. The advantage of the dye-sensitized solar cell (DSSC) is utilized indoor fluorescent light illumination, but the output power of DSSC is not high enough. Therefore, we propose the titanium dioxide (TiO2) – magnet beads (MBs) – reduced graphene oxide (rGO) composited photoanode (TMGP) to enhance the photovoltaic conversion efficiency under low Illumination. It under the indoor light condition can maintain photovoltaic conversion efficiency (PCE) by about 14%.

II. EXPERIMENTAL DETAILS

A. MATERIALS

The organic sensitized dye was N719, which purchased from Solaronix Aubonne (Switzerland). The platinum target (99.99% purity) for sputtering was purchased from Ultimate Materials Technology Co., Ltd. (Taiwan). The electrolyte contained with methoxy propionitrile (MPN), iodine (I2), 4-Tert-Butylpyridine (TBP), lithium iodide (LiI), and 1-propyl-2,3-dimethylimidazolium iodide (DMPII). The titanium tetrachloride (TiCl4) and acetylacetone (AcAc) were Sigma-Aldrich, (United States).

B. PREPARATION OF THE COLLOIDAL PASTE

The MBs-TiO2 spin paste had consisted of 1.5 g P25 TiO2 powder, 0.1 ml acetic acid, 3 ml deionized water (D.I. water) and 0.125 mM MBs. The rGO-TiO2 doctor blade paste was prepared as follows: Firstly, the 5 mg rGO powder put in 4 ml D.I. water that the solution into an ultrasonic oscillator for one hour. The purpose was dispersed rGO powder evenly in the solution. After that, 2 g TiO2 powder and 0.4mL absolute alcohol were added into the rGO solution. Finally, the rGO-TiO2 solution mixture was stirred for one day to obtain rGO-TiO2 colloid paste. Besides, we prepared pure TiO2 colloid paste without MBs and rGO powder [17].

C. FABRICATION OF THE PHOTOANODE FOR DYE-SENSITIZED SOLAR CELL

The fluorine-doped tin oxide (FTO) glass (7 Ω/cm2) was cleaned by acetone, ethanol, and D.I. water, respectively. The TiCl4 stock solution was diluted with D.I. water to the 40 mM concentration TiCl4 treatment solution via the ice bath method. Then the cleaned FTO was immersed into 40 mM TiCl4 solution at ~ 70 °C for the half-hour to make the TiO2 compact layer then annealing at 450 °C for the half-hour at the furnace. The working layer was based on the TiO2 compact layer by using the spin method and doctor blade method that the photoanode annealing at 450 °C for the half-hour. Finally, the annealed photoanode used TiCl4 treatment and annealing for a half-hour. Annealing the photoanode film at an ambient temperature of 450 °C enables the removal of organic impurities in the photoanode film and strengthens the contact between nanoparticles within the film.

D. INSTRUMENTATION

The photocurrent-voltage curve measurements were conducted under Xe lamp solar simulator (MFS-PV-Basic-HMT, Taiwan) with the sunlight intensity of 100 mW/cm2 and indoor fluorescent light source (T5) and light decay filter. Besides, we used light filter 80%, 50%, 30%, and 10% to investigate the DSSCs performance of different sunlight intensities. The Nyquist plot was used to measure the frequency range of 1 MHz to 50 Hz in the dark at a potential of 0.7 V.

III. RESULTS AND DISCUSSION

A. PHOTOVOLTAIC PERFORMANCES OF THE DSSC WITH TiO2 AND TMGP

The current-voltage curves (J-V) of DSSC based on TiO2 and TMGP, the photovoltaic conversion efficiency (PCE) is enhanced by 31.75 % from 5.20 % to 6.85 %, as shown in Fig. 1. This improvement in photovoltaic conversion efficiency is due to the enhancement of short-circuit current density (Jsc) by magnetic beads (MBs) and reduced graphene oxide (rGO). First, the MBs provides another charge transfer path, which can improve the charge transfer characteristics in the TiO2 film and reduce the electron recombination opportunity. Besides, the rGO is acted as a bridge to accelerate the excited-electron from the conduction band (CB) of titanium dioxide (TiO2) to the CB of FTO. This role of rGO can reduce the dark reaction between the excited-electron and oxidized-dye molecule [18], [19], [20], [21], [22], [23].

Moreover, the equivalent circuit for DSSC in this study is exhibited in Fig. 2, and it would not exhibit in electrochemical Impedance Spectroscopy (EIS) measurement as below. It composes of Rs, R1, R2, C1, and C2. First, the Rs indicates the resistance between FTO substrate and wire. Besides, Z1 indicates high-frequency impedance of interface between electrolyte and counter electrode, which is the
first semicircle at the left-hand side. An equivalent circuit, $Z_1$ indicates the parallel connection between $R_1$ and $C_1$, and $R_1$ is the interface resistance between the counter electrode and electrolyte. Additionally, $Z_2$ is the frequency impedance of interface between the electrolyte and the active layer (photoanode), which is the second semicircle at the right-hand side. An equivalent circuit, the $Z_2$ is the parallel connection between $R_2$ and $C_2$, and $R_2$ is the interface resistance between the active layer (photoanode) and electrolyte. Additionally, the DSSC is operated in direct current, that the capacitance can be ignored.

It can be seen from Fig. 2 that $R_2$ is reduced from 11.4 $\Omega$ to 6.8 $\Omega$ because MBs provide another charge transfer path for the photoanode for better electron transfer, and the rGO can increase the amount of dye in the photoanode and reduce charge recombination. Because of the above properties, MBs and rGO enable more excited electrons to be transported to external circuits. Finally, the $J_{SC}$ current can be enhanced [24], [25]. Furthermore, the details of MBs and rGO were shown in our previous research [17].
TABLE 1. Photovoltaic parameters of DSSC based on TMGP under different light intensities.

| Intensity(mW/cm²)/ | $V_{OC}$ (V) | $J_{SC}$ (mA/cm²) | F.F. (%) | PCE (%) |
|-------------------|--------------|-------------------|---------|---------|
|        (Lux)      |              |                   |         |         |
| 100/ 683 K        | 0.75         | 15.73             | 59.33   | 6.85    |
| 80/ 546 K         | 0.74         | 13.45             | 62.37   | 7.76    |
| 50/ 341 K         | 0.72         | 8.65              | 65.63   | 8.17    |
| 30/ 204 K         | 0.71         | 5.75              | 68.10   | 9.27    |
| 10/ 68 K          | 0.69         | 1.98              | 72.29   | 9.88    |

TABLE 2. Photovoltaic parameters of DSSC based on TiO$_2$ under different light intensities.

| Intensity(mW/cm²)/ | $V_{OC}$ (V) | $J_{SC}$ (mA/cm²) | F.F. (%) | PCE (%) |
|-------------------|--------------|-------------------|---------|---------|
|        (Lux)      |              |                   |         |         |
| 100/ 683 K        | 0.63         | 518.4             | 75.52   | 14.08   |
| 80/ 546 K         | 0.61         | 458.3             | 74.38   | 14.13   |
| 50/ 341 K         | 0.61         | 303.2             | 75.76   | 14.58   |
| 30/ 204 K         | 0.60         | 157.4             | 74.29   | 14.69   |
| 10/ 68 K          | 0.53         | 51.9              | 68.86   | 8.52    |

TABLE 3. Photovoltaic parameters comparison of photovoltaic parameters of DSSC with other literature [26].

| Intensity(mW/cm²)/ | $V_{OC}$ (V) | $J_{SC}$ (mA/cm²) | F.F. (%) | PCE (%) |
|-------------------|--------------|-------------------|---------|---------|
|        (Lux)      |              |                   |         |         |
| 100/ 683 K        | 0.65         | 15.01             | 57.13   | 5.66    |
| 80/ 546 K         | 0.64         | 12.51             | 58.31   | 5.84    |
| 50/ 341 K         | 0.62         | 8.40              | 59.44   | 6.29    |
| 30/ 204 K         | 0.61         | 5.47              | 60.68   | 6.86    |
| 10/ 68 K          | 0.59         | 1.71              | 61.36   | 5.88    |

TABLE 4. Photovoltaic parameters of DSSC based on TMGP in series and parallel.

| Connection | Active area (cm²) | $V_{OC}$ (V) | $J_{SC}$ (mA/cm²) | F.F. (%) | PCE (%) |
|------------|------------------|--------------|-------------------|---------|---------|
| Series     | 1.00             | 0.73         | 15.50             | 58.55   | 6.61    |
| Series     | 1.00             | 1.41         | 7.65              | 57.28   | 6.16    |
| Series &Parallel | 2.00          | 1.42         | 15.23             | 55.04   | 5.93    |
| Series     | 2.56             | 1.46         | 13.08             | 59.13   | 4.48    |
| Series     | 2.56             | 0.73         | 28.16             | 59.14   | 4.35    |

B. PHOTOVOLTAIC PERFORMANCE OF THE DSSCs IN SERIES-PARALLEL MODULES

Fig. 3 (a) and (b) show the photovoltaic conversion efficiency (PCE) of DSSC based on TiO$_2$ and TMGP under low illumination. The TMGP photoanode of DSSC is enhanced 44.23 %, from 6.85 % to 9.88 %, and the TiO$_2$ photoanode of DSSC is enhanced 24.29 %, from 5.23 % to 6.49 %. In addition, the open-circuit voltage ($V_{OC}$) is decreased from 0.75 V to 0.69 V, which can be attributed to the decrease in the amount of excited-electron. Because the $V_{OC}$ is the difference between Fermi-level of active layer and redox potential of electrolyte. Besides, the amount of excited-electrons are reduced with decline of light intensity. Similarly, because the amount of excited-electron is decreased, the $J_{SC}$ current is also reduced. The enhancement in photovoltaic conversion efficiency is due to the increase in fill factor (F.F.). The fill factor is increased from 59.33 % to 72.29 %. However, the better PCE of DSSC can be obtained under lower illumination than indoor fluorescent, which are due to fewer amount of excited-electrons. Moreover, the higher improvement in photovoltaic conversion efficiency under low illumination can be obtained from DSSC based on TMGP. Because the amount of excited-electrons is little, the decrease of dark reaction is helpful for the improvement of photovoltaic conversion efficiency of DSSC under low illumination.

In the result with Table 1 and Table 2, the PCE of DSSC based on TiO$_2$ decreased from 11.39 % to 5.50 % as the light intensity was decreased from 1.75 mW/cm² to 0.22 mW/cm². In addition, the $J_{SC}$ dropped from 462.5 µA/cm² to 46.8 µA/cm² as the light intensity was decreased from 1.75 mW/cm² to 0.22 mW/cm². Moreover, the $V_{OC}$ of DSSC based on TiO$_2$ decreased from 0.62 V to 0.49 V as the light
intensity was decreased from 1.75 mW/cm$^2$ to 0.22 mW/cm$^2$. Because the dye molecule has a better response with indoor fluorescent lamp, the $V_{OC}$ of 1.75 mW/cm$^2$ was higher than that of 10 mW/cm$^2$. In addition, the $F.F.$ was decreased from 69.49% to 52.74%, while the light intensity was decreased from 1.75 mW/cm$^2$ to 0.22 mW/cm$^2$. Moreover, the result in the PCE decreased from 11.39% to 5.50%. Photovoltaic conversion efficiency decreased from 11.39% to 5.50% while light intensity was decreased from 1.75 mW/cm$^2$ to 0.22 mW/cm$^2$.

In Table 3, we compare DSSCs under different light intensities with other literature. Chou et al. in [26], the DSSC in the IGZO retardation structure. The PCE is enhanced by 21.20%, from 5.66% to 6.86%. In Table 4, we compare the DSSC module with other literature. Chou et al. in [27], the DSSC in parallel connection and series connection are fabricated by silver paste connection. The $I_{SC}$, $V_{OC}$, $PCE$ of series connection DSSC are 13.08 mA/cm$^2$, 1.46 V and 4.58%. The $I_{SC}$, $V_{OC}$, $PCE$ of parallel connection DSSC are 28.16 mA/cm$^2$, 0.73 V and 4.35%.

According to previous literature [26], [27], the reduced dark reaction for photoanode of DSSC is a key point to enhance photovoltaic performances, but there are a few literature to apply the reduced dark reaction on DSSC. Moreover, DSSCs in series-parallel has not been investigated, and it has the potential to develop to drive the device.

**IV. CONCLUSION**

In summary, the properties of DSSCs with MBs and rGO in series-parallel have been investigated. The DSSCs with MBs and rGO can obtain higher photovoltaic conversion efficiency under low illumination, which is due to the retardation of dark reaction. Because the amount of excited-electrons is few under low illumination, a decrease in reverse recombination is a key point to increase the photovoltaic conversion. MBs provides another charge transfer path, which can improve the charge transfer. Besides, the rGO is acted as a bridge to accelerate the excited-electrons from the conduction band of FTO to the conduction band of titanium dioxide (TiO$_2$).

In other words, rGO can retard the dark reaction between the excited-electrons and oxidized-dye molecules. The advantage of the dye-sensitized solar cell (DSSC) is utilized indoor fluorescent light illumination, but the output power of DSSC is not high enough under indoor fluorescent light illumination. Moreover, the output power can be enhanced by series and parallel.

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