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Grid type dye-sensitized solar cell module with carbon counter electrode

Won Jae Lee^{a,*}, Easwaramoorthi Ramasamy^{a,b}, Dong Yoon Lee^a, Jae Sung Song^a

 ^a Electric and Magnetic Devices Research Group, Korea Electrotechnology Research Institute, P.O. Box 20, Changwon 641-120, Republic of Korea
^b University of Science & Technology, Daejeon 305-333, Republic of Korea

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Abstract

To realize low cost, high-performance dye-sensitized solar cell (DSSC) technology on industrial scale, large area grid type DSSC module has been prepared on silver grid-embedded transparent conducting glass substrate. Commercial titanium dioxide (TiO₂) and carbon powders were employed to make working and counter electrodes, respectively. Under simulated solar light (AM 1.5, P_{in} : 100 mW cm⁻²), 5 cm × 5 cm size carbon counter electrode module with an active area of 11.2 cm² shows V_{OC} : 0.730 V, I_{SC} : 118 mA, FF: 0.55 with 4.23% active area efficiency, which is comparable to 5.26% of platinum counter electrode module. © 2007 Elsevier B.V. All rights reserved.

Keywords: Solar cell; Counter electrode; Metal grid; Module; Efficiency

1. Introduction

Over 10% energy conversion efficiency in state of the art, laboratory scale dye-sensitized solar cell (DSSC) has attracted more fundamental as well as commercial interest [1,2]. Industrial researchers have been taking considerable efforts to scale up this low cost, high-performance technology in the form of transparent windows [3], stand-alone power generator [4]. However, commercial entry of this type solar cell is so far hindered by the poor stability associated with electrolyte leakage, degradation of dye at elevated temperature and dissolution of platinum counter electrode in highly corrosive iodide/tri-iodide (I^{-}/I_{3}^{-}) redox couple electrolyte [5]. In recent years, more number of works has been carried with the aim of improving the both, stability and performance. Notable progress is achieved in the field of stable sensitizer [6], low volatile electrolyte [7] and new counter electrode materials. Among the group of alternative counter electrodes studied so far, carbon emerged as suitable material for large size DSSCs, owing to good corrosion resis-

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tance in I^-/I_3^- redox electrolyte and cheapness. Relatively low catalytic activity of carbon towards I_3^- reduction may compensate by enhancing the active surface area with optimum pore size [8–10].

Up scaling the DSSC technology also rapidly improved since past few years. Unlike conventional silicon cells, thin film solar cells including DSSCs are fabricated on transparent conductive oxide (TCO) substrate and their limited conductivity imposes performance limitation, especially in large area devices [11]. To avoid such resistive losses, DSSC modules are fabricated by connecting few millimeter wide stripe type devices in Z or W type interconnection [12]. Series-monolithic type, where entire module is fabricated on single substrate through successive layer coating also intensively studied for inexpensive roll-to-roll production process [11,13]. On the other hand, high performance, large area DSSC may be prepared by using metal grid-embedded TCO substrate. High aperture ratio and simple production methods make this module more favorable in high current applications. However, the presence of corrosive electrolyte in DSSC urges the need of overcoat layer for metal grids [14].

In this respect, here we investigated the applicability of nanosize carbon powder as counter electrode material for large size DSSCs. Using such carbon counter electrode, $5 \text{ cm} \times 5 \text{ cm}$ size

^{*} Corresponding author. Tel.: +82 55 280 1643; fax: +82 55 280 1590. *E-mail address:* wjlee@keri.re.kr (W.J. Lee).



Fig. 1. Schematic cross-section view of grid type DSSC module with carbon counter electrode.

grid type DSSC module with comparable energy conversion efficiency and stability was fabricated.

2. Experimental

2.1. Metal grid-embedded substrates

Working electrodes and counter electrodes were prepared on F-doped SnO₂ glass (sheet resistance: $10 \Omega/\Box$) substrates. Using silver paste (Electro-Science Laboratories Inc.), grid lines with the dimension of 5 µm × 300 µm (height × width) were screen printed on previously cleaned FTO glass substrate. After leveling at room temperature for 10 min, Ag lines were dried at 180 °C to avoid damage due to follow-up active layer printing step.

2.2. Working and counter electrodes

Screen printable TiO₂ paste was prepared from commercial powder (Deagussa, P25) as described in our previous work [15]. A screen with 200 mesh is used to obtain mechanically rigid, ~10 μ m thick TiO₂ layer after sintering at 500 °C for 1 h. Dye sensitization is carried out by immersing the sintered electrodes in 0.3 mM dye solution (N719, Solaronix) in ethanol for 24 h. Counter electrodes were prepared using a paste, consists of carbon powder (primary particle size: 40 nm) in organic binder added water on pre-drilled, Ag grid-embedded substrate by doctor blading method. After drying at room temperature for 10 min, electrodes were subsequently sintered at 250 °C for 1 h in air. Platinum counter electrodes were prepared by using hexachloroplatinic acid-based screen printable paste, as demonstrated in our previous work [16].

2.3. Module preparation and current-voltage measurement

DSSC module consists of four stripe type TiO₂ layers (width \times length: 7 mm \times 40 mm) on Ag grid-embedded substrate was prepared by staking the counter electrode on top of the sensitized TiO₂ electrode. Patterned surlyn foil (thickness: 80 µm) was kept between two electrodes as a spacer come overcoat layer for Ag grid lines. Using hot press, the structure was sealed and inter-electrode space was filled with liquid electrolyte consists of 0.5 M LiI, 0.05 M I₂ and 0.5 M 4-tertbutylpyridine in acetonitrile. The electrolyte filling holes were sealed by surlyn and glass cover. Fig. 1 schematically shows the cross-section view of assembled grid type DSSC module with carbon counter electrode. In order to compare the performance of the carbon counter electrode DSSC, platinum counter electrode DSSC also prepared at identical condition. Current-voltage performance of the device is characterized in both dark and simulated AM 1.5 solar illumination (Oriel, Model: 91160, 300 W Xenon arc lamp) using computer controlled digital source meter (Keithley, Model: 2400). The incident light intensity was adjusted to desired level by using neutral density filter and photometer (International light, Model IL1400).

3. Results and discussion

3.1. Microstructural characterization

Field emission scanning electron microscope (FE-SEM) images of TiO₂ and carbon film on FTO glass substrate were shown in Fig. 2. The mesa-porous structure of the TiO₂ as well as carbon film was clearly seen from FE-SEM images. In order to enhance the catalytic activity of carbon towards triiodide reduction, we used nano-size carbon powder with particle size of 40 nm as shown in Fig. 2b. Sintering the carbon film at 250 °C leads to the formation of interconnected network with optimum porous structure. Comparable performance of our carbon counter electrode may attribute to the high surface area with porous nature, which makes I_3^- ion diffusion favorable throughout the film and thus entire surface area is available for I_3^- reduction [17].

3.2. Current-voltage characteristics

The performance of carbon counter electrode was evaluated through measuring current-voltage curve of the device in both



Fig. 2. FE-SEM images of TiO₂ (a) and carbon (b) film on FTO glass substrate.

Counter electrode	$\frac{P_{\rm in}~(\rm mWcm^{-2})}{}$	$V_{\rm OC}$ (V)	$\frac{I_{\rm SC}~(\rm mA)}{}$	Fill-factor (%)	Efficiency (%)	
					Active area	Total area
Platinum	53	0.730	54.5	0.72	4.83	2.16
	100	0.750	119.0	0.66	5.26	2.36
Carbon	53	0.700	53.9	0.63	4.00	1.79
	100	0.730	118.0	0.55	4.23	1.90

Table 1 I-V parameters^a of grid type DSSC module under different illumination conditions

^a Values are average of two cells.



Fig. 3. I-V curve of 5 cm × 5 cm size, grid type DSSC module under simulated solar light (P_{in} : 100 mW cm⁻², AM 1.5). Solid line represents the device with carbon counter electrode and broken line represents the device with platinum counter electrode. Active area of the device is 11.2 cm^2 .

dark and range of illumination condition. Fig. 3 presents the I-V performance of grid type module, consists of four stripe type TiO₂ layers with an active area of 11.2 cm². At present manual spacer pattern and position techniques limit the module's aperture ratio (active area/total area) around 45% in 5 cm × 5 cm size substrate. To have a good comparison, performance of platinum counter electrode module is also shown in the same figure. Under one sun illumination, both modules show similar short-circuit current (I_{SC}) and open-circuit voltage (V_{OC}). Here comparable I_{SC} may attribute to high surface area and thus enhanced

catalytic performance of carbon counter electrode. However, low fill-factor of carbon counter electrode module, especially at one sun condition, brings down the active area energy conversion efficiency to 4.23% (total area efficiency: 1.90%). Large number of interconnection between carbon particles increased counter electrode's contribution to internal resistance of the device and hence low fill-factor [18]. On the other hand, platinum counter electrode module shows 5.26% active area efficiency (with respect to total area: 2.36%). The detailed behavior of carbon and platinum counter electrode DSSC module for two different levels of illumination is shown in Table 1.

3.3. Stability

Few micron thick carbon layer, made up of nano-size powder increases the concern that loosely bounded particle may detach from the counter electrode side, enhance the I_3^- reduction near TiO₂ electrode and hence low performance. Fig. 4 shows the short-term evaluation of *I–V* parameter of carbon counter electrode DSSC module up to 9 weeks. During the test, modules were stored in dark at room temperature and current–voltage characterization was carried out at particular interval under one sun illumination condition. During the first 4 weeks of study V_{oc} of module is gradually increased and stabilized afterwards. However, I_{sc} is gradually decreased throughout the test except first week and the same trend is observed in platinum counter electrode module also (not shown). As shown in Fig. 4b, carbon counter electrode module retaining 92% of its initial energy conversion efficiency after 9 weeks of aging. During the sealing



Fig. 4. Stability of grid type DSSC module with carbon counter electrode (a) open-circuit voltage V_{oc} and short-circuit current I_{sc} and (b) energy conversion efficiency η and fill-factor FF. For comparison, efficiency of Pt module is also shown.



Fig. 5. Photograph of grid type DSSC module with platinum (a) and carbon (b) counter electrodes.

process, both sides of the module were heated at 100 °C for few seconds. This may cause dye degradation and hence drop in I_{sc} .

3.4. Appearance

Moderate transmittance in the visible region and multifarious color [19] makes DSSCs more attractive in building integrated photovoltaic technology (BIPV). Fig. 5 shows the visual appearance of grid type DSSC module with platinum and carbon counter electrode over a background image. Transparent nature of Pt counter electrode and sealing material makes clear appearance of background image (see Fig. 5a). In the case of carbon counter electrode module, moderate transmittance can be achieved via optimization of aperture ratio, as shown in Fig. 5b.

4. Conclusion

A carbon counter electrode DSSC module, with 4.23% overall light to electric energy conversion efficiency was realized on 5 cm \times 5 cm size, silver line embedded FTO glass substrate. The comparable performance of carbon counter electrode with platinum is attributed to high surface area associated with nanosize carbon powder. Our promising result shows that carbon can be successfully used in commercial scale DSSCs, where transparency is not a main issue.

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