EFFECT OF FILM THICKNESS OF REDUCED GRAPHENE OXIDE COUNTER ELECTRODES ON PHOTOVOLTAIC PROPERTIES OF DYE SENSITIZED SOLAR CELLS

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INTRODUCTION

Dye-sensitized solar cell (DSSC) is an electrochemical device that converts solar energy into electricity, and this device is first reported by O'Regan and Grätzel in 1991 (O'Regan, 1991). A typical structure of the DSSC includes three major components: a dye adsorbed TiO_2 photoanode, a redox electrolyte, and a counter electrode (CE). Out of these, the CE plays a key role in regulating the performance of DSSC as a catalyst for the redox couple regeneration and electron collection from the external circuit (Wu, 2012). Generally, platinum (Pt) deposited on Fluorine-doped tin oxide (FTO) conductive glass is used as a CE due to its excellent electro-catalytic activity for the triiodide reduction, high conductivity, and good chemical stability. Nevertheless, Pt is a kind of non-renewable and limited natural resource, which limits large-scale applications. However, due to the high cost and chemical degradation of Pt in the corrosive iodine environment demand more cost-effective, electrolyte resistance, and earth-abundant alternatives for DSSC applications (Jiang, 2010).

To achieve similar CEs with low cost, high electronic conductivity and comparable catalytic effects for tri-iodide reduction, various potential alternative materials have been investigated to replace Pt. Some of them are conducting polymers such as polyaniline, polypyrrole, etc., metal sulphides such as nickel sulphide and cobalt sulphide and carbon-based materials such as graphene, reduced graphene oxide. Among them, carbon-based materials attract much attention as substitutes for Pt due to their low cost. Carbon materials such as graphite (Li, 2015), graphene oxide (Mahmoud, 2019), reduced graphene oxide (Mohan, 2019), and graphene (Roy-Mayhew, 2010), etc. are preferred alternative CE materials. The preparation method of carbon-based CEs is always associated with some problems such as low adhesion to the conducting substrate, thereby unsatisfactory redox performances of carbon materials and low charge-transfer ability. To overcome these problems, we introduced a spray method with very low usage of binders to bind carbon material on conducting substrate. In this study, Reduced Graphene Oxide (RGO) was used to fabricate low- cost CEs for DSSCs and CE film thickness was optimized according to the photovoltaic properties of DSSCs.

METHODOLOGY

CEs were prepared using RGO by spray technique. For that, spray solution was prepared by mixing different amounts of RGO, acetic acid, Triton X-100, and

ethanol and sonicated for 1 hour before spray. Then this mixture was sprayed on

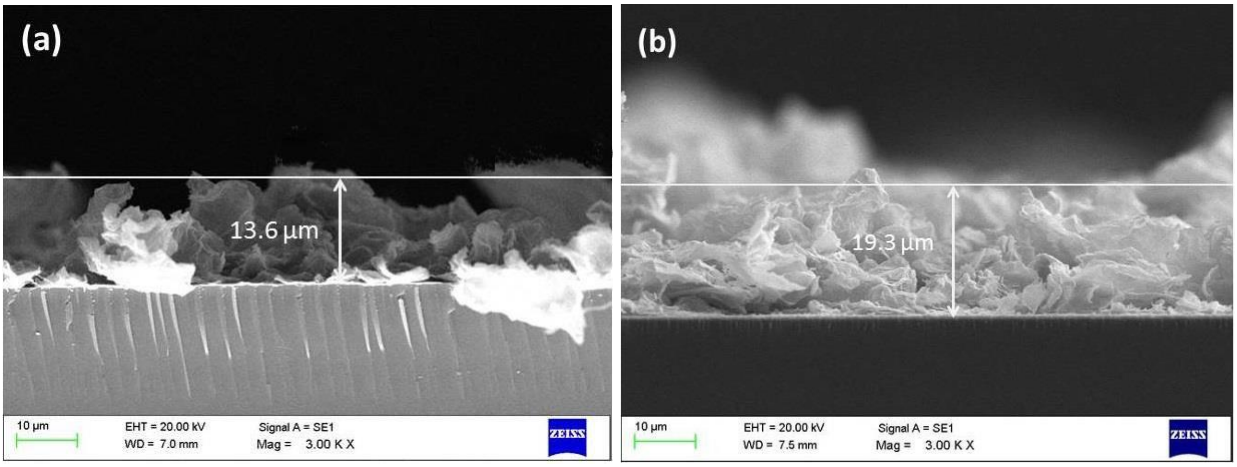
FTO glass substrate while heating the FTO glass substrate at 100 ^{o}C. The CE film thickness was varied by using different amounts of RGO as 5 mg, 10 mg, 15 mg, and 20 mg coated on the surface area of 6 cm^2. Finally, they were sintered at 250 ^{o}C for 45 minutes, in a muffle furnace. The liquid electrolyte used in this study contains 0.6 M 1-methyl 3- propyl imidazolium iodide (PMII) ionic liquid, 0.03 M Iodine, 0.1 M guanidinium thiocyanate, 0.5 M 4-tert-Butylpyridine (TBP) and acetonitrile.

Two layers of TiO_2 were made on the conducting glass substrate which was used as the photoanode (Kumari, 2017). To prepare the first layer, P90 TiO_2 powder was used by preparing a paste mixing with 0.1 M HNO_3. Then this paste was coated on the cleaned FTO glass substrate by spin coating technique.

The second layer of TiO_2 was coated on the first layer using P25 TiO_2 powder. The creamy paste was prepared by P25 TiO_2 powder mixing with 0.1M HNO_3, triton X-100, and PEG 2000. The paste was doctor bladed on the first layer and sintered at 450 °C for 45 minutes to obtain a porous TiO_2 layer. After cooling down to room temperature, they were dipped in Ru N719 dye for 24 hours. DSSC was fabricated by sandwiching the liquid electrolyte in between dye attached TiO_2 photo-anode and RGO CE with the active cell area of 0.16 cm^2.

RESULTS AND DISCUSSION

Figure 1 shows the SEM images of the cross-section of prepared RGO based CEs corresponding to different film thickness. It can be observed based on these images that the film thickness is not uniform due to the fluffy and wrinkled nature of RGO sheets. However, the film thickness was increased gradually with the increase of the RGO amount used to prepare CEs from 5 mg to 20 mg coated on the total surface area of 6 cm^2.



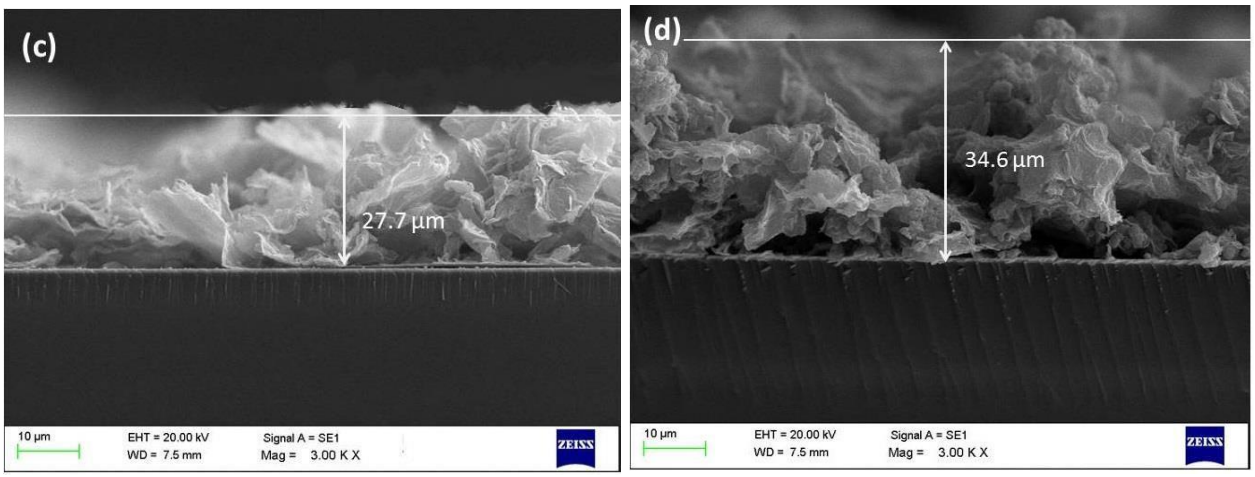


Figure 1. SEM images of the cross-sections of RGO CEs with different film thicknesses (a) 13.6 µm, (b) 19.3 µm, (c) 27.7 µm, and (d) 34.6 µm.

The current density-voltage (*J-V*) characteristics of the fabricated DSSCs employing different counter electrodes were studied under light irradiation of 100 mW cm^{-2}. To investigate the effect of CE film thickness on the performance of DSSCs, a series of RGO CEs were prepared by varying the used RGO amount as 5 mg, 10 mg, 15 mg, and 20 mg, respectively. The total area of CEs was kept fixed as 6 cm^2. The best DSSC performance was exhibited by 15 mg of RGO used CEs corresponding to RGO film thickness of 27.7 µm as shown in SEM image (Figure 1(c)). The highest efficiency of 4.70 % was observed for this optimized RGO CE-based DSSC. Figure 2(a) shows the J-V curve of DSSCs with different thicknesses of RGO CEs and Pt CE, and Table 1 shows the CE film thickness information and photovoltaic parameters of DSSCs.

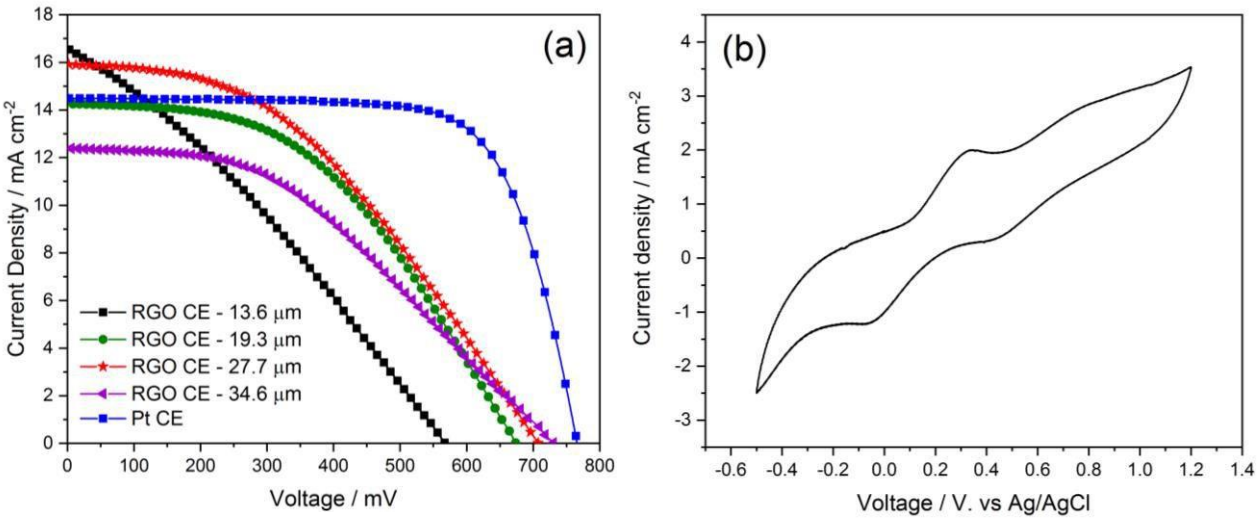


Figure 2. (a) Current density-voltage characteristics graphs of DSSCs with Pt CE and RGO CEs with different thicknesses, (b) Cyclic voltammetry graphs of RGO, in a three-electrode arrangement using a 50 mV s^{-1} scan rate.

Table 1. Photovoltaic parameters of DSSCs based on RGO CEs with different film thickness and comparison them based on Pt CE.

RGO amount used (mg)	**CE film thickness according to SEM images (μm)**	**J_{sc} / mA cm^{-2}**	**V_{oc}/ mV**	**FF %**	**Efficiency %**
5	13.60	16.60	567.0	30.42	2.86
10	19.30	14.30	673.6	46.47	4.48
15	27.70	15.90	707.3	41.79	4.70
20	34.60	12.40	730.4	41.05	3.72
Pt CE	-	14.47	775.1	69.80	7.82

Cyclic voltammetry measurements were obtained to investigate the electrochemical behaviors of RGO CE. The characteristic plots for optimized CE were recorded using an iodide-based liquid electrolyte with a scan rate of 50 mV s^{-1} by applying a sweep potential from -0.5 V to 1.2 V. Ag/AgCl electrode was used as reference electrode and Pt rod was used as the counter electrode. Prepared RGO CE was used as the working electrode with an active surface area of 1.0 cm^2. Figure 2(b) shows the cyclic voltammetry analysis graphs for RGO CE. Two distinctive sets of peaks related to oxidation and reduction reactions were observed for optimized RGO CE confirming the excellent electrocatalytic activity towards the I^-/I_3^- redox reaction of the electrolyte.

CONCLUSIONS/RECOMMENDATIONS

We have successfully fabricated RGO counter electrodes by spray method for applications in dye-sensitized solar cells. This simple and low cost preparation method provides the formation of a porous structure with improved electrocatalytic effect for triiodide ion reduction at the CE and efficient ionic mobility for iodide/triiodide ions in the electrolyte medium. The best energy conversion efficiency of DSSCs is obtained for RGO based CE with a film thickness of 27.7 μm (efficiency of 4.70 %). This value is comparable to the efficiency of 7.82% for Pt CE-based DSSC operating under similar conditions.

ACKNOWLEDGMENTS

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