



Performance enhancement effects of dispersed graphene oxide sponge nanofillers on the liquid electrolytes of dye-sensitized solar cells

Shanmuganathan Venkatesan^a, Elmer Surya Darlim^a, I-Ping Liu^a, Yuh-Lang Lee^{a, b, *}

^a Department of Chemical Engineering, National Cheng Kung University, No.1 University Road, Tainan, 70101, Taiwan

^b Research Center for Energy Technology and Strategy, National Cheng Kung University, No.1 University Road, 70101, Tainan, Taiwan

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ABSTRACT

A graphene oxide sponge (GOS) is prepared and utilized as a nanofiller (NF) of an iodide liquid electrolyte for dye-sensitized solar cell (DSSC) application. The concentration effects of the GOS NFs on the conductivity of the electrolyte, and the performance of the DSSCs are studied. The results indicate that the inclusion of GOS NFs markedly enhances the conductivity of the liquid electrolyte. The electrochemical impedance spectroscopy (EIS) analysis shows that the presence of GOS NFs may increase the recombination resistance (R_{ct}) at the photoelectrode/electrolyte interface and, furthermore, decreases the charge transfer resistance at the Pt counter electrode/electrolyte interface (R_{pe}). Therefore, the current density (J_{sc}), open circuit potential (V_{oc}), and fill factor (FF) of the DSSCs can be improved by controlling the concentration of GOS NFs. In this study, the DSSC with 0.50 wt% GOS NFs can achieve the highest energy conversion efficiency of 9.44%, which is higher than that obtained for the corresponding cell without GOS NFs (8.84%).

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1. Introduction

Dye-sensitized solar cells (DSSCs) have been considered to be a suitable alternative to replace classical silicon solar cells owing to their low production cost, simple fabrication, and relatively high energy conversion efficiency [1–4]. In DSSCs, the electrolytes play a key role in determining cell efficiency and stability. Various kinds of electrolytes, such as conventional organic solvent-based liquid electrolytes, gelled liquid electrolytes, ionic liquids, inorganic and organic hole transport materials, polymer gel electrolytes have been developed thus far for DSSC applications [5,6]. As we know, liquid electrolytes are mostly used as the transport medium for DSSCs owing to their high energy conversion efficiency in comparison to other electrolytes [6–9]. The components of the liquid electrolyte comprise the organic solvent, redox couple, and the additive [10–12]. Among these, the redox couple is a key component of the electrolyte involved in dye-regeneration as well as charge transport between the electrodes and the electrolytes in

DSSCs [13–15]. Since the beginning of DSSC development, the iodide/triiodide (I^-/I_3^-) is the most investigated redox system for DSSCs [16,17]. This is because of the unique characteristics of the I^-/I_3^- redox couples, including good solubility in solvents, high conductivity and good penetration ability into the nanostructured TiO_2 films [16], [18–20]. Although the I^-/I_3^- redox couple shows good performance in DSSCs, there are some adverse features restricting its applications in DSSCs. The most critical features are the recombination reaction between the electrons and iodide ions, as well as the mismatch between the redox potential of a typical sensitizer and that of the I^-/I_3^- redox couple, which affects the performance of the DSSCs [5,14,16,21]. Various methods, such as using cobalt redox couples, utilizing dyes with more hydrophobic chains, optimizing the morphology of the TiO_2 , employing carbon-based counter electrodes, using cations with higher charge density, and so on, have also found to be beneficial to solve these problems [9], [22–27].

In the literature, several studies focused on the utilization of nanosized inorganic nanoparticles as nanofillers (NFs) of electrolytes for DSSC applications [28]. The addition of various kinds of NFs in the electrolytes is a simple, effective way to increase the photovoltaic performance of DSSCs. Owing to their high versatility

* Corresponding author. Department of Chemical Engineering, National Cheng Kung University, No.1 University Road, Tainan, 70101, Taiwan.

E-mail address: ylllee@mail.ncku.edu.tw (Y.-L. Lee).

in structural, mechanical and electrochemical properties, carbonaceous materials are considered as effective NFs for DSSC electrolytes [28–42]. Among the carbonaceous NFs, two dimensional (2D) GO was utilized as the gelator of acetonitrile (ACN) and 3-methoxy propionitrile (MPN) based iodide liquid electrolytes [40–41]. However, the presence of GO sheets in these electrolytes triggered several problems. First of all, the larger size of the 2D GO sheets decreased the conductivity of the electrolytes because they are not ionically conductive and may hinder the charge transport [40]. Secondly, the cell using GO sheets had a high charge transfer resistance at the electrolyte/counter electrode interface (R_{pt}) due to partial coverage of Pt-surface by GO sheets [41]. It was reported that, the presence of carbonaceous NFs in the electrolytes promoted the generation of polyiodides [32,34,36], leading to the increase of the electrolyte conductivities [43]. However, the higher polyiodide concentration also enhanced the recombination reactions that significantly reduced the open circuit potential (V_{oc}) of the cells [32,34,36,44]. Therefore, the overall performance of the DSSCs was affected. To solve these problems, the utilization of other carbonaceous NFs are required.

In this study, we investigated the role of a graphene oxide sponge (GOS) dispersed in a liquid electrolyte in an attempt to achieve an improvement in the photovoltaic characteristics of DSSCs using an I^-/I_3^- redox couple. A GOS has a hierarchal three-dimensional structure (3D) with amphiphilic surfaces [45–47]. It also has a high surface area and a large pore volume, as well as high thermal, chemical and electrochemical stability in organic solvents. Owing to these features, GOS NFs in liquid electrolyte can function as a solid-state electron mediator and mobile counter electrode (CE), which is able to increase ionic mobility and catalytic reactions of the I^-/I_3^- redox couple, as well as to decrease the formation of polyiodides. Therefore, the current density (J_{sc}), V_{oc} , and fill factor (FF) of DSSCs can be improved. In this study, the effects of various amounts of the GOS NFs on the properties of liquid electrolytes as well as on the performance of the DSSCs are extensively studied. The GOS NFs were beneficial in preparing a high performance DSSC with an efficiency higher than that of its liquid counterpart.

2. Experimental

2.1. Materials

Iodine (I_2 , 99.99%), lithium iodide (LiI, 99%), 4-*tert*-butylpyridine (tBP, $C_9H_{13}N$, 96%), guanidine thiocyanate (GuSCN, $CH_5N_3 \cdot HSCN$, 99%), hydrogen peroxide (H_2O_2 , 30%) and sodium nitrate ($NaNO_3$, 99%) were obtained from Sigma Aldrich. Sulfuric acid (H_2SO_4 , 97–98%), ethanol (EtOH, 99%), potassium permanganate ($KMnO_4$) were purchased from J. T. Baiker. Graphite powder and 1,3-dimethylimidazolium iodide (DMII, $C_5H_9N_2I$, 99%) were received from Bay carbon Inc and Alfa Aesar, respectively. PST-400C and PST-18NR titanium dioxide pastes were purchased from JGC C & C (JGC Catalysts and Chemicals Ltd., Japan). The N719 ruthenium dye, di-tetrabutylammonium *cis*-bis(isothiocyanato)bis(2,2'-bipyridyl-4,4'-dicarboxylato)ruthenium(II), ($C_{58}H_{86}N_8O_8RuS_2$, 99%) was obtained from Solaronix. All of these chemicals were used as received.

2.2. Preparation of graphene oxide

The graphene oxide (GO) was synthesized from graphite powder using a modified Hummer's method [49]. Following typical preparation methods, 1.5 g graphite powder (300 mesh) was added to 37 ml of H_2SO_4 (98%) in a reactor. Then, 4.50 g of $KMnO_4$ and 0.75 g of $NaNO_3$ were added gradually to the mixture under constant and vigorous stirring, and the temperature of the mixture was kept below $10^\circ C$. Then, the mixture was stirred using an

ultrasonication bath for 2 h at $35^\circ C$ to obtain a homogeneous mixture. Then, 70 ml of de-ionized water was added, and the mixture was stirred at $90^\circ C$ for 15 min. Subsequently, another 190 ml water was added to the mixture to reduce the temperature of the mixture. Finally, a solution of 3.6 ml of H_2O_2 (30%) was added to this mixture. The suspension was separated by centrifuging the mixture at 8000 rpm for 10 min three times. After that, the GO suspension was centrifuged at 1400 rpm for 10 min to obtain muddy GO. Then, the muddy GO was dried at $60^\circ C$ for 24 h to obtain the GO sheets.

2.3. Preparation of the GO sponge

The GOS was produced by freeze-drying a GO dispersion in water for 48 h according to the reports published in the literature [47]. Briefly, a conical vial containing the GO dispersion in water was placed in a cold source (liquid nitrogen) maintained at a constant temperature. The temperature was maintained for a few hours ($\sim 2-3$ h) until the GO solution was completely solidified. Then, the solidified sample was kept in a freeze-dryer and vacuum dried for 48 h, producing the GO sponges. The GOS was characterized by using various physiochemical techniques. The surface morphology of GOS was investigated by using scanning electron microscope (SEM). Both picture and SEM image of GOS was given in Fig. 1. The results obtained in these experiments were consistent with reports in the literature [44–48].

2.4. Preparation of photoanodes

TiO_2 photoanodes were prepared via screen printing (SP) of the TiO_2 pastes on a transparent conductive substrate (TCS). Briefly, fluorine doped tin oxide (FTO, TEC 7, 2.2 mm thick, $7 \Omega sq^{-1}$) as the TCS was washed with a course of detergent solution, acetone, and de-ionized water. After drying the TCS, a mixture of 10 ml of pure EtOH and 30 μl 0.01 M of $Ti(i-Pro)_4$ was coated on the TCS as a single layer and dried at $50^\circ C$ for 30 min. This pretreatment process created a chemical bond between the TiO_2 thin film and the TCS. Then, PST-18NR and PST-400C TiO_2 pastes were coated on the TCS as the main layer (8.0 μm) and the scattering layer (4 μm), respectively. Finally, the TiO_2 /TCS was heated at $500^\circ C$ for 30 min and allowed to cool to $80^\circ C$. Then, the TiO_2 /TCS electrode was immersed in N719 dye in ethanol (0.5 mM) at room temperature

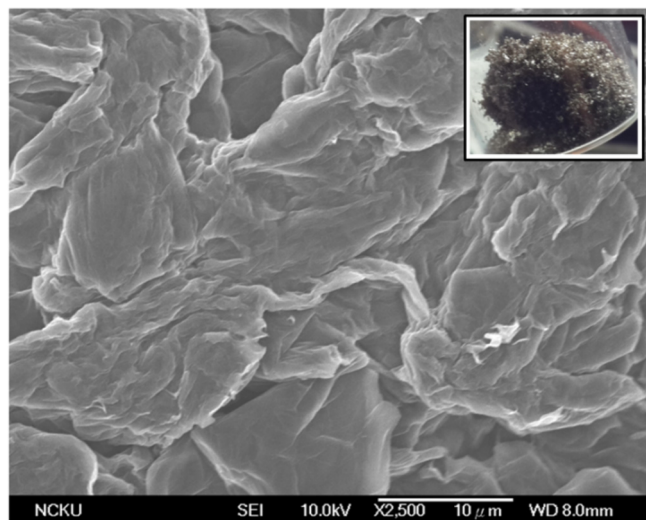


Fig. 1. Scanning electron microscope image of GOS. The inset shows a picture of as prepared GOS. (A colour version of this figure can be viewed online.)

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