



Performance studies of ZnO and multi walled carbon nanotubes-based counter electrodes with gel polymer electrolyte for dye-sensitized solar cell

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ABSTRACT

Counter electrode (CE) is one of the major component which determine the energy conversion efficiency of the dye-sensitized solar cells (DSSCs). Here in, five types of CEs using two different materials were fabricated. Three types are zinc oxide (ZnO) based while other two are multi-walled carbon nanotubes based CEs. The thickness and resistance per cm^2 of each CE were measured. The gel polymer electrolyte based on polyacrylonitrile polymer, sodium iodide salt, 1-Hexyl-3-methyl-imidazolium iodide ionic liquid was also prepared. The conductivity studies revealed that the highest ionic conductivity of gel polymer electrolyte was 6.72 mS cm^{-1} upon incorporation of 100 wt% 1-Hexyl-3-methyl-imidazolium iodide ionic liquid. This gel polymer electrolyte was sandwiched between commercial TiO_2 photo-anode and different types of CE to fabricate the DSSCs. The J - V characteristic curves of the DSSCs were obtained and the DSSC characterization parameters were determined. The efficiencies achieved using ZnO and MWCNT-based CEs were ranging from 0.46% to 7.07%. The results demonstrate that ZnO and multi-walled carbon nanotubes are good candidates of CEs for DSSC application.

1. Introduction

Dye-sensitized solar cells (DSSCs) are one of the new generation of solar cells which draw noticeable attention by the researchers due to its distinct advantages such as low-cost benefit and facile fabrication [1,2]. They are promising devices for clean energy conversion in utilizing the renewable solar energy [3–5]. A general DSSC consists of semiconductor thin film coated with a single layer of sensitizing dye, a redox species of iodide/tri-iodide (I_3^-/I^-) electrolyte, and a counter electrode with catalytic property [6,7]. One of the important components in a DSSC which affects its performance is the CE. A CE play vital role in collecting electrons generated by the dye and reducing the I_3^-/I^- redox species [7,8]. Therefore, the most preferred CE must be able to possess excellent catalytic property and high ionic conductivity [3]. The most popular material for CE which fulfils these conditions is Pt. But the high cost, scarcity and the degradable performance due to corrosive electrolyte are the main hindrances for the commercialization Pt as CE in DSSC. Therefore, alternatives to replace to the Pt are need to search [2,9,10].

Large scale investigations have been performed to replace the Pt from the counter electrode with other highly efficient electrode

materials [11]. Conducting polymers such as poly(3,4-ethylenedioxythiophene) (PEDOT), polyaniline and polypyrrole remain attracted considerable interest as a counter electrode due to their high catalytic activity to perform I^-/I_3^- redox reaction [12–14]. Recently, sulfide, nitrides, and carbides materials demonstrated excellent performance as counter electrodes in DSSC by decent reduction of I_3^- in iodine-based electrolytes [3,15,16]. One of the efficient alternative for CE preparation is carbon-based material. Generally, they are low in cost, electrically conductive, highly corrosion resistant and environmental friendly [17,18]. Carbon black [19], carbon nanotube [20], fullerene and graphene [21] are some of the carbon-based materials that are largely used to investigate their performance for CE in DSSCs.

CNTs are unique nano-sized tubular shaped allotropes of carbon which provide outstanding properties like large surface area, high electrical and thermal conductivity, good mechanical properties and chemical stability. A multi-walled carbon nanotubes (MWCNTs) shows a “bamboo-like” structure consists of several graphene sheets arranged co-axially together [3,8,10,22]. Since the catalytic property of MWCNT are excellent, they can catalyse the redox species in the electrolyte to certain extent [3].

On the other hand, ZnO is given attention due to its excellent

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catalytic activity, diverse morphologies, corrosive resistant and large surface area. Its wider band gap (3.37 eV) makes it transparent in the electromagnetic visible spectrum [23–25]. It is low-cost and bears high electron mobility compared to the TiO₂ based DSSCs. Due to its large surface area, ZnO favours the adsorption of dye molecules. This metal oxide shows a good possibility for application in DSSCs [3,26].

In the present study, we report the fabrication of CE for DSSC using MWCNTs and ZnO on fluorine doped tin oxide (FTO) substrates. Moreover, GPE is also prepared by using polyacrylonitrile (PAN) as host polymer, ethylene carbonate (EC) and propylene carbonate (PC) as solvent, sodium iodide (NaI) salt, 1-Hexyl-3-methyl-imidazolium iodide (HMII) ionic liquid, and iodine (I₂). The composition, crystallinity and conductivity was evaluated using, FTIR, XRD and EIS respectively. The efficiencies of the fabricated DSSCs using different CEs are measured and compared with the commercial Pt CE.

2. Methodology

2.1. Materials

2.1.1. Preparation of gel polymer electrolyte (GPE)

The materials used for GPEs were PAN, EC, PC, NaI salt, HMII ionic liquid, and iodine (I₂). Both PC and EC were stirred at 100 °C for 10 min, followed by adding NaI and iodine into the solution and stirred for further 30 min. Finally, appropriate amount of HMII ionic liquid and PAN was added. The weight percentage (wt%) of HMII ionic liquid with respect to the polymer was varied (20, 40, 60, 80, and 100 wt%) for optimization

2.1.2. Counter electrode preparation

2.1.2.1. Multi-walled carbon nanotube based counter electrode. To prepare the paste for counter electrode, a 50 mg of MWCNT was added to 1 mL of ethyl alcohol (EA), stirred and sonicated for 30 min. 2.5 mg of poly(vinylpyrrolidone-co-vinylacetate) (PVP) was stirred and sonicated with the mixture for 30 min. This paste was coated in two ways, that is, by applying two drops on a FTO glass using dropper (labelled as CE-D); and by spin-coating on the FTO glass at 1000 rpm for 2 s and 2350 rpm for the following 60 s (labelled as CE-S). Lastly, the samples were sintered at 450 °C for 30 min.

2.1.3. Zinc oxide (ZnO) based counter electrode

The first ZnO based CE (labelled as ZX-0) was prepared by mixing 10 drops of dimethyl sulfoxide (DMSO) with 0.2 g of grounded ZnO nanoparticle. The paste was coated on FTO glass by doctor blade method and sintered at 450 °C for 30 min. The second CE (labelled as ZX-1) was prepared by adding 1 drop of Triton X-100 to the paste used for ZX-0 before coating. The last ZnO-based CE (ZE-S) was prepared by sonicating 1 mL of EA with 50 mg of grounded ZnO for 30 min, followed by sonicating 2.5 mg of PVP for the same duration. The paste was spin-coated and sintered at 450 °C for 30 min.

2.1.4. Dye-sensitized solar cell fabrication

To prepare porous photo-electrode, double layers of TiO₂ paste was coated on the FTO glass and sintered at 450 °C for 30 min. For the first layer, 0.5 g of grounded TiO₂ (P90) was mixed with 2 mL of nitric acid (pH = 1) and spin coated onto the FTO glass. The two-step spin coating was performed at 1000 rpm for 2 s and then 2350 rpm for 60 s. For the second layer, 2 mL of nitric acid (pH = 1) was added into the 0.5 g grounded TiO₂ (P25). After that 0.1 g of Carbowax and one drop of Triton X-100 were added to the paste. This paste was coated using doctor blade method on the first layer and sintered at 450 °C. Then, it was soaked in the commercial N719 dye solution for 24 h. The prepared gel polymer electrolyte (GPE) was sandwiched between the photo-electrode and counter electrode.

2.1.5. Characterization methods

2.1.5.1. Electrochemical impedance spectroscopy (EIS). The ionic conductivity and temperature-dependent ionic conductivity studies on the GPE were performed using Hioki 3532–50 LCR HITESTER spectrometer. The ionic conductivity (σ) of the GPE was computed by the following equation:

$$\sigma = \frac{L}{R_b A} \quad (1)$$

where σ is the ionic conductivity (S cm⁻¹), L denotes the thickness of the film (cm), R_b represents the bulk resistance (Ω) and A is the area of blocking electrodes (cm²). For temperature dependent ionic conductivity study was performed from room temperature followed by 30–100 °C in the step of 10 °C.

2.1.6. Fourier transform infrared (FTIR) spectroscopy

The composition of the GPEs were analyzed by Thermo Scientific, Nicolet iS10 FTIR spectrometer. The studies carried out in the wave-number ranging from 4000 to 650 cm⁻¹ with 1 cm⁻¹ resolution in transmittance mode.

2.1.7. X-ray diffraction (XRD)

XRD Siemens D 5000 diffractometer (40 kV, 40 mA) was employed to obtain the crystallinity of the GPEs, in the setting of Cu K α radiation, and wavelength of 1.540600 Å for 2 θ range of 5–80° at room temperature.

2.1.8. Field emission scanning electron microscopy

Nanoscale images of the coating materials of the fabricated electrodes were taken by JEOL JSM-7600F field emission scanning electron microscopy (FESEM) fitted with EDX and elemental mapping was employed to analyse the surface morphology.

2.1.9. Properties of counter electrodes

The resistance of each CE across an area of 1 cm² was measured using DT-830B Digital Multimeter. The thickness of CE coatings was taken using KLA Tencor P-6 surface profiler.

2.1.10. Characterization of DSSC

The GPE based DSSCs were analyzed under sun simulator (ABET Technologies Solar Simulator), in which the samples were illuminated by a 1000 W m⁻² white light to obtain the J - V characteristics curves for each samples.

3. Results and discussion

The ionic conductivity of each GPEs was studied using EIS. Fig. 1 displays the Cole-cole plot of GPE with 100 wt% of HMII with respect to PAN at room temperature. The bulk resistance (R_b) of this sample was easily identified as 18.52 Ω as demonstrated in Fig. 1.

Fig. 2 represents the variation of ionic conductivity of the GPEs against the addition of HMII ionic liquid content. It is evident from the Fig. 2 that ionic conductivity of GPE at room temperature was increased as the contents of HMII ionic liquid was increased. The value of ionic conductivity improved from 4.95 mS cm⁻¹ to 6.72 mS cm⁻¹ by raising the contents of ionic liquid from 20 wt% to 100 wt% respectively. The increment of ionic conductivity was owing to the numerous charge carriers provided by the elevated contents of ionic liquid which can further enhance the photovoltaic performance of the system [27].

Furthermore, the positive complex ions of HMII are able to adsorb on the TiO₂ photo-electrode surface. This adsorption can lead in the negative shift of the conduction band of TiO₂. The open-circuit voltage, V_{oc} is related to the conduction band of the photo-electrode and the potential of the redox electrolyte, the addition of HMII ionic liquid in GPE can increase the V_{oc} , and improve the performance of DSSC [28]. Since, the GPE with HMII content of 100 wt% with respect to PAN

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