



Influence of carbon nanotubes in gel electrolyte on photovoltaic performance of ZnO dye-sensitized solar cells

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ABSTRACT

In this work, the poly(ethylene glycol) (PEG) gel polymer electrolyte was used to replace the liquid electrolyte in dye-sensitized solar cells (DSSCs) in order to overcome the issues of evaporation, leak of liquid, and stability. However, the PEG gel polymer electrolyte-ZnO based cells have low current density and fill factor. Then, the effect of addition of multi-walled carbon nanotubes (MWCNTs) in different wt% into the PEG gel polymer electrolyte was also investigated. Current density–voltage characteristics and electrochemical impedance spectroscopy showed that use of the MWCNTs with the PEG gel polymer electrolyte increases current density and fill factor. Highest efficiency of 0.75% was achieved with 5 wt% of the MWCNTs in the PEG gel polymer electrolyte. The enhancement of efficiency can be attributed to the suppression of charge transfer resistance at the ZnO photoelectrode/electrolyte interface, whereas the proper introduction of MWCNTs effectively decreases the charge recombination as confirmed by electrochemical impedance spectra. In case of stability, the devices sustained for 1006 h also maintained over 85% of its initial performance.

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1. Introduction

Dye-sensitized solar cells (DSSCs) are considered to be one of the promising alternatives to the existing silicon based-solar cells due to ease of fabrication and low cost. The high light-to-electricity conversion efficiency make them an ideal candidate for the next generation of solar cells [1,2]. Current generation from these devices relies on the electron injection from photo-excited state of the sensitizer dye into the conduction band of the wide band gap semiconductor electrode, then the HOMO level of dye is subsequently restored by the electron donation from electrolyte material consisting of iodide (I^-) and triiodide (I_3^-) which is derived from I^- and I_2 by ion exchange in a reduction/oxidation reaction [3]. The energy conversion efficiency exceeding 12% has been achieved under air mass 1.5 irradiation (100 mW/cm^2) in TiO_2 DSSCs using the cosensitization of porphyrin dye used in conjunction with $\text{Co}^{(II/III)}$ tris(bipyridyl)-liquid based redox electrolyte [4]. Recently,

the research on ZnO as an alternative photoelectrode in DSSCs has been intensively carried out because its band gap, electron affinity, and electron injection efficiency are similar to those of TiO_2 . To date, ZnO nanostructures-based DSSCs have achieved promising power conversion efficiency values of over 7.14% [5]. The major drawbacks of these cells, however, are the corrosion and evaporation of the electrolyte solvent as well as photodegradation of the attached photoelectrode. These downsides severely affect the long-term operation of DSSCs performance [6]; therefore, efforts have been made worldwide to develop more stable DSSCs.

In order to improve stability of the DSSCs, inorganic semiconductors were employed in DSSCs as electrolyte. It has been shown that the inorganic p-type semiconductor, which employed in solid state DSSCs, can improve the stability. There is, however, a trade-off in terms of low conversion efficiency because the mobility of the inorganic material is lower compared to that of the liquid electrolyte. This can be explained by the poor quality of the contact between photoelectrode and material interface and the limitation of charge transfer in the p-type semiconductor [7]. Therefore, to improve the stability, it is necessary to replace the liquid electrolyte with the quasi-solid state electrolyte using polymer matrix chains, such as gel polymer electrolyte, containing the solution of I^-/I_3^- redox couple [8–10]. Moreover, the gel polymer electrolyte

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can eradicate the contact interface problem because it possesses liquid like viscosity at high temperature. Hence, it is possible for the gel polymer electrolyte to penetrate into a mesoscopic space of the photoelectrode [8]. Although this approach would solve the problem, the charge-transfer kinetic via the gel polymer medium of I^-/I_3^- redox couple is limited at room temperature. Therefore, raising the ionic mobility of the gel polymer electrolyte is necessary to improve the cell performance. To further improve the cell efficiency, many efforts so far have focused on the inorganic semiconductor nanofiller that can be applied to reduce the charge transfer resistance of the gel polymer electrolyte [11,12].

Multi-walled carbon nanotubes (MWCNTs) can be used as a nanofiller which improve a charge transport in the medium due to their distinct characteristics such as electronic properties, ballistic charge transport, high mechanical strength, thermal stability, and catalytic property for the reduction of triiodine to iodine ion [13–17]. With these prominent points, MWCNTs can be regarded as a candidate material for various applications. Thus, herein, we demonstrated that adding MWCNTs into the gel polymer electrolyte of ZnO DSSCs can improve the energy conversion efficiency of the cell.

Inspired by the important role that the gel polymer electrolyte overcomes not only the stability concern but also the contact interface problem, in this study, the gel polymer electrolyte was prepared by using poly(ethylene glycol) (PEG) as polymer matrix, $LiI + I_2$ dissolved in propylene carbonate as electrolyte solution, and MWCNTs as nanofiller. Properties of the prepared gel polymer electrolyte with different amount of MWCNTs were characterized by the field emission scanning electron microscopy (FE-SEM) and the Fourier transform infrared spectroscopy (FT-IR). Finally, the obtained MWCNTs gel polymer electrolyte was applied between two electrodes of ZnO DSSCs and the complete ZnO-based cell with the MWCNTs gel polymer electrolyte was investigated in detail by the electrochemical impedance spectroscopy (EIS) experiment and the energy conversion efficiency measurement.

2. Experimental

2.1. MWCNTs gel polymer electrolyte preparation

Liquid electrolyte containing LiI and I_2 was used to create reduction and oxidation species in polar solvent of propylene carbonate ($C_4H_6O_3$, $102.09 \text{ g mol}^{-1}$). First, the liquid electrolyte was prepared by mixing 0.7 M LiI and 0.07 M I_2 in propylene carbonate. Next, the gel polymer electrolyte was prepared by adding high molecular weight PEG ($20,000 \text{ g mol}^{-1}$, 41 wt\%) to the liquid electrolyte. In addition, the MWCNTs (reported elsewhere [6,13]) as a nanofiller were then added into this gel polymer electrolyte. Finally, the mixture of the liquid electrolyte together with PEG and MWCNTs was subsequently sonicated at 60°C for 30 min in order to retrieve a homogeneous mixture.

2.2. ZnO DSSC fabrication

DSSCs were fabricated on conduction glass (fluorine-doped SnO_2 glass, FTO glass with sheet resistance of $8 \Omega \text{ cm}^{-2}$). ZnO powder (Aldrich, 99.9%) was also mixed in PEG solution to form ZnO powder paste. Then, the paste was coated onto the FTO substrate as a photoelectrode of DSSCs by doctor blade technique. Thereafter, the ZnO films on FTO were sintered at 400°C for 1 h. The area and the thickness of the films were about $0.5 \times 2 \text{ cm}^2$ and $30 \mu\text{m}$, respectively. The ZnO photoelectrodes were immersed in an organic dye Eosin-Y sensitizer ($C_{20}H_6Br_4Na_2O_5$, in 100 cm^3 of ethanol, Sigma Aldrich) for 1 h at room temperature. A platinum counterelectrode was prepared by dropping a $10 \mu\text{L}$ of a platinum chloride solution

(0.5 mM hydrogen hexachloroplatinate (IV) hydrate, $\text{Cl}_6\text{H}_2\text{Pt.aq}$, in acetone solution, Sigma Aldrich) on the FTO substrate and then sintered at 550°C for 1 h. The dye-loaded ZnO photoelectrode and the Pt counterelectrode were assembled into a sealed device using a hot-melted double layer parafilm ($50 \mu\text{m}$ -thicks per sheet). Subsequently, the PEG gel polymer electrolyte was melt on heat bath at 60°C and then loaded into the gap between the active surfaces of both electrodes.

2.3. Measurement of charge transfer resistance

To analyze the interfacial charge transfer resistance (R_{ct}) on each region of the PEG gel polymer electrolyte ZnO DSSCs, electrochemical impedance spectroscopy (EIS, Hioki model 3522-50) and sourcemeter potentiostat (Keithley model 2611) were utilized in the frequency range $0.02\text{--}10^4 \text{ Hz}$. Using EIS, the interfacial charge transfer resistance R_{ct} can be obtained by analyzing an equivalent circuit of the DSSCs. EIS spectra were recorded under dark condition with a voltage and current source operated at about a forward bias open circuit voltage. The obtained EIS spectra were then fitted using Z view software to obtain parameters in the equivalent circuit.

In order to understand the influence of each ion on the cell performance, the obtained EIS parameters were then used to calculate the ionic conductivities (σ) of the electrolytes using the following equation [12]:

$$\sigma = F \sum_i |z_i| u_i C_i = \frac{l}{R_{ct} \cdot A}, \quad (1)$$

where F , z_i , u_i , and C_i are the Faraday constant, the ionic charge number, the ionic mobility, and the ionic concentration, respectively; l is the length of the space between the photoelectrode and the counterelectrode, R_{ct} is the interfacial charge transfer resistance, and A is the an active area of the cell.

2.4. Measurement of energy conversion efficiency

Cell parameters of the PEG gel polymer electrolyte ZnO DSSCs were determined using an AM1.5 solar simulator with the radiant power of 100 mW cm^{-2} for all illuminations. The intensity of the incident beam was calibrated with a standard Si solar cell. Current–voltage characteristics were measured with a computer controlled current–voltage source. The energy conversion efficiency (η) was then calculated through to the following equation [18]:

$$\eta = \frac{J_{sc} V_{oc} FF}{P_{in}}, \quad (2)$$

where J_{sc} , V_{oc} , FF , P_{in} are current density measured at short circuit, the open circuit voltage, the fill factor of the cell, and the intensity of the incident light, respectively. The fill factor (FF) can be calculated from [18]:

$$FF = \frac{P_{max}}{J_{sc} V_{oc}}, \quad (3)$$

where the maximum power P_{max} is obtained as the product of the current density and voltage at the maximum output power of the cell.

3. Results and discussion

3.1. Characterization of MWCNTs gel polymer electrolytes

The MWCNTs gel polymer electrolytes with the variation of MWCNTs concentrations show homologous and slurry with high viscosity. The morphology of the MWCNTs gel polymer electrolyte

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