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Journal of Photochemistry Photobiology A:Chemistry

Journal of Photochemistry and Photobiology A: Chemistry 181 (2006) 333-337

www.elsevier.com/locate/jphotochem

Quasi-solid state dye-sensitized solar cells-based gel polymer electrolytes with poly(acrylamide)-poly(ethylene glycol) composite

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Received 31 August 2005; received in revised form 3 November 2005; accepted 15 December 2005 Available online 24 January 2006

Abstract

A novel gel polymer electrolyte based on poly(acrylamide)–poly(ethylene glycol) composite and binary organic solvent was prepared. And the highest ionic conductivity of the gel polymer electrolyte was 1.94 mS cm^{-1} under the ambient atmosphere. The polymer gel electrolyte based on the composite of poly(acrylamide)–poly(ethylene glycol), the binary organic solvent of ethylene carbonate and propylene carbonate and the additive of 4-*tert*-bytylpyridine has been employed to fabricate quasi-solid state dye-sensitized solar cell with an overall conversion efficiency of approximately 3% under irradiation of 60 mW cm⁻².

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Keywords: Quasi-solid state dye-sensitized solar cells; Gel polymer electrolytes; Poly(acrylamide)-poly(ethylene glycol) composite; Ionic conductivity

1. Introduction

Dye-sensitized solar cells (DSSC) based on sensitization of TiO₂ nanocrystalline by photoexcited dye molecules have been investigated intensively because of its lower cost and potential alternatives to traditional photovoltaic device [1–3]. light-to-electrical energy conversion efficiencies of DSSC based on liquid electrolytes using organic compound, such as acetonitrile, propylene carbonate and ethylene carbonate as solvent and iodide/triiodide (I^-/I_3^-) redox couple as electrolyte have reached 10–11% under irradiation of AM 1.5 [2–7]. However, this type of liquid-junction cell remains some problems including low long-term stability which caused by organic solvent evaporation and leakage of liquid electrolytes, high temperature instability and difficulties in sealing the devices [8].

To overcome these problems, much effort has been made to replace the liquid electrolytes with solid or quasi-solid type charge transport materials [9–12]. Compared with other kinds of charge transport materials, the gel polymer electrolytes have some advantages including high ionic conductivities which are

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achieved by "trapping" a liquid electrolyte in polymer cages formed in a host matrix, good contacting and filling properties with the nanostructured electrode and counter electrode. Therefore, the gel polymer electrolytes have been attracting intensive attention. Up to the present, several types of gel electrolytes based on different kind of polymers have already been used in quasi-solid state dye-sensitized solar cells [12–14].

Poly(acrylamide) possess carbonyl group and amine group on its molecular chain [15], also poly(ethylene glycol) have many hydroxyl group on its molecular chain. It is expected that the interaction between sensitized dye and the matrix of gel polymer can be improved based on the hydrogen bond interaction between the carbonyl group on sensitized dye and the carbonyl group, amine group and hydroxyl group on matrix of gel polymer electrolyte. On the other hand, the ionic conductivity of the gel polymer electrolyte can be enhanced according to the complexation from carbonyl group, amine group and hydroxyl group on poly(acrylamide) and poly(ethylene glycol) to K⁺ ions in electrolyte. Consequently, the overall conversion efficiency of the DSSC can be enhanced. In this paper, using poly(ethylene glycol) as both reactant and plasticizer, a novel homogeneous poly(acrylamide)-poly(ethylene glycol) composite without phase separation was synthesized. Using the composite as matrix, and 4-tert-bytylpyridine as

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additive, the binary organic compounds ethylene carbonate and propylene carbonate as solvent, a gel polymer electrolyte with quasi-solid state was prepared. Further, a dye-sensitized solar cell was fabricated by sandwiching the gel polymer electrolyte.

2. Experimental

2.1. Materials

Poly(ethylene glycol) with average molecular weight 400 (PEG-400), acrylamide monomer, ammonium persulfate, potassium iodide, iodine, ethylene carbonate (EC), propylene carbonate (PC) and γ -butyrolactone (γ -BL) were all A.R. grade and all purchased from Xilong Chemicals. PEG-400 was dried at 120 °C for 12 h before used. Other reagents were used without further treating before using.

Conducting glass plates (FTO glass, Fluorine doped tin oxide over-layer, sheet resistance $8 \Omega \text{ cm}^{-2}$, purchased from Hartford Glass Co., USA) were used as a substrate for precipitating TiO₂ porous film on and were cut into $2 \text{ cm} \times 1.5 \text{ cm}$ sheets. Sensitizing dye *cis*-[(dcbH₂)₂Ru(SCN)₂] was purchased from SOLARONIX, SA.

2.2. Synthesis of the poly(acrylamide)–poly(ethylene glycol) in situ composite

Poly(acrylamide)-poly(ethylene glycol) in situ composite was synthesized by adding 3 g PEG-400 into 6 g acrylamide monomer. The mixture was heated at 70-75 °C to melt the monomer and mix two components homogeneously. The mixture was marked as A. A polymerization initiator (ammonium persulfate, 1 wt.% of acrylamide monomer) and PEG-400 (1 g) were mixed and stirred until the mixture dissolved entirely at room temperature, and the mixture was marked as B. Under vigorous stirring and keeping a temperature at 70-75 °C on a water bath heater, the mixture B was added into the mixture A slowly, the polymerization reaction took place and a homogeneous mixture formed, the mixture was marked as C. It is noticeable that as soon as the polymerization reaction begins, the mixture C should be put out of the water bath heater and down to room temperature to prevent implosion because the reaction is exothermic reaction. When the mixture C became a viscous gel in ambient environment, it was heated again at 60-65 °C for 30 min to complete polymerization reaction. After that, the poly(acrylamide)-poly(ethylene glycol) in situ composite was synthesized.

2.3. Preparation of gel polymer electrolyte

A mixture organic solvent was made by mixing ethylene carbonate (EC), propylene carbonate (PC), γ -butyrolactone (γ -BL). Potassium iodide [KI = 10 wt.% of total weight of the composite and the mixture solvent] and iodine (I₂ = 10 wt.% of KI) were dissolved in the mixture organic solvent to form a liquid electrolyte. Suitable amount of poly(acrylamide)–poly(ethylene glycol) composite was added into the liquid electrolyte under continuous stirring at room temperature to form a gel polymer electrolyte with quasi-solid state.

2.4. Assembling of the quasi-solid state dye-sensitized solar cell

Nanoporous TiO₂ film was manufactured by following procedure. Tetrabutyl titanate (20 ml) was rapidly added to distilled water (200 ml) and a white precipitate was formed immediately. The precipitate was filtered using a glass frit and washed three times with 100 ml of distilled water. The filter cake was added to nitric acid aqueous solution (0.1 M, 200 ml) under vigorous stirring at 80 °C until the slurry became a translucent blue–white liquid. The resultant colloidal suspension was autoclaved at 200 °C for 12 h to form milky white slurry. The resultant slurry was concentrated to 1/4 of its volume, then PEG-20000 (10 wt.% slurry) and a few drops of emulsification regent of Triton X 100 were added to form a TiO₂ colloid.

A conducting glass sheet (FTO) was immersed in an isopropanol solution for 48 h to remove any impurities. A plastic adhesive tape was fixed on the four sides of conducting glass sheet to restrict the thickness and area of TiO₂ porous film. The TiO₂ colloid was dropped on the FTO glass plate by using a doctor scraping technique. The process was done for three times to form a thick TiO₂ film about 6–8 μ m. The TiO₂ film was treated with 50 mM TiCl₄ aqueous solution in ambient environment for 12 h and washed with distilled water. Finally, the TiO₂ porous film was solidified and sintered by firing the conducting glass sheet at 450 °C in air for 30 min.

After cooled to $80 \,^{\circ}$ C, the TiO₂ film was immersed in a 2.5×10^{-4} M absolute ethanol solution of *cis*-[(dcbH₂)₂Ru(SCN)₂] for 24 h to absorb the dye adequately, the other impurities were washed up with anhydrous ethanol and dried in moisture-free air. After that, a TiO₂ porous film electrode absorbed dye was prepared.

Quasi-solid state dye-sensitized solar cell was assembled by dropping a drop of gel polymer electrolyte into the aperture between the TiO_2 porous film electrode (anode electrode) and a Pt plated conducting glass sheets (cathode electrode, prepared by electrodepositing). The two electrodes were clipped together and a cyanoacrylate adhesive was used as sealant to prevent the electrolyte solution from leaking.

2.5. Measurements

Poly(acrylamide)–poly(ethylene glycol) composite thermal stability was measured by a SDT2960 simultaneous DSC–TGA device (USA TA Instrument) over a temperature range of 30-600 °C at a heating rate of 10 °C min⁻¹. The ionic conductivity of gel polymer electrolytes was measured by using model DDB-6200 digitized conductivity meter (Shanghai Reici Instrument Factory, China). The instrument was calibrated with 0.1 M KCl aqueous solution prior to experiments.

The photovoltaic test of quasi-solid state dye-sensitized solar cells was carried out by measuring the J-V character curves under irradiation of white light from a 100 W xenon arc lamp (XQ-500W, Shanghai Photoelectricity Device Company, China)

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