

Influence of ionic additives NaI/I₂ on the properties of polymer gel electrolyte and performance of quasi-solid-state dye-sensitized solar cells

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Abstract

The ionic additives NaI/I₂ in polymer gel electrolyte not only provide cations, but also affect the liquid electrolyte absorbency of the poly(acrylic acid)–poly(ethylene glycol) hybrid, which results in the change of ionic conductivity of polymer gel electrolyte and the photovoltaic performance of quasi-solid-state dye-sensitized solar cell. With the optimized components of liquid electrolyte containing 0.5 M NaI, 0.05 M I₂, 0.4 M pyridine, 70 vol.% γ -butyrolactone and 30 vol.% *N*-methylpyrrolidone, a 4.74% power conversion efficiency of quasi-solid-state dye-sensitized solar cell was obtained under 100 mW cm⁻² (AM 1.5) irradiation.

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Keywords: Liquid electrolyte absorbency; Poly(acrylic acid)–poly(ethylene glycol) hybrid; Polymer gel electrolyte; Quasi-solid-state dye-sensitized solar cell

1. Introduction

Dye-sensitized solar cell (DSSC) consists of a dye-coated mesoporous nanocrystalline TiO₂ photoelectrode, platinum modified counter electrode and a thin electrolyte layer sandwiched between the above two electrodes. Since the prototype of DSSC reported in 1991, a record power conversion efficiency about 11% has been achieved with organic-solvent electrolyte [1,2]. Since then, DSSCs have received great attention and broad research over the world due to their high power conversion efficiency with low production cost. However, DSSCs with organic-solvent electrolytes show a substantial disadvantage of poor long-term stability for their practical uses, owing to the leakage and volatilization of encapsulated organic solvents. Thus, work has been done to replace organic-solvent electrolytes with p-type inorganic semiconductors or hole-conducting materials but currently with moderate success [3–6]. Recently, DSSCs using gel electrolytes prepared by adding low molecular gelators or polymer materials showed comparable efficiencies to the cells using organic-solvent electrolytes [7–15]. The following advantages of polymer gel electrolytes lead to their broad application in fabricating quasi-solid-state

DSSCs. Firstly, they show high ionic conductivity by “trapping” volatile organic solvents in polymer cages formed in polymer matrix. Secondly, they can obtain good contacting and filling properties with nanoporous TiO₂ film and counter electrode. Furthermore, their fabrication procedures are simple. However, some kinds of polymer gel electrolytes mainly based on van der Waals force or other physical effects between polymer chains to solidify liquid electrolytes [16–18]. They are thermo-reversible and unstable. So a rigorous sealing procedure is still need for fabricating good long-term stability of DSSCs using these polymer gel electrolytes.

Poly(acrylic acid)–poly(ethylene glycol) (PAA–PEG) hybrid is a kind of efficient liquid electrolyte absorbent, which can absorb large amount of Lewis basic mixed solvents, such as γ -butyrolactone (GBL) and *N*-methylpyrrolidone (NMP) based on acid–basic interaction between the hybrid and mixed solvents [19]. Polymer gel electrolyte based on the hybrid is thermo-irreversible and stable due to the 3D network structure of the swollen hybrid [20,21]. It is found that the liquid electrolyte absorbency of the hybrid is a key parameter in ionic conductivity of polymer gel electrolyte and photovoltaic performance of quasi-solid-state DSSCs [19–21]. Ionic additives such as LiI, NaI, KI and I₂ are providers of cations in polymer gel electrolyte and sources of iodide/triiodide redox couples for regeneration of oxidized dyes. The additives also influence the liquid electrolyte absorbency of the hybrid by the interactions among alkali

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metal ions and carbonyl or ether groups in the hybrid based on complexation [20]. Therefore, it is necessary to investigate the influence of ionic additives on the properties of polymer gel electrolyte and performance of quasi-solid-state DSSCs. In this paper, NaI is selected as the source of I^- due to its relatively high solubility in GBL and NMP mixed solvents and less sensitivity to water. The optimized components of 70 vol.% GBL and 30 vol.% NMP mixed solvents are used in the polymer gel electrolyte [19,22].

2. Experimental

2.1. Materials

PEG with average molecular weight 1000, acrylic acid monomers, ammonium persulfate, sodium pyrosulfite, titanium iso-propoxide, nitric acid, glacial acetic acid, terpineol, ethyl cellulose, ethanol, γ -butyrolactone, *N*-methylpyrrolidone, pyridine (PY), sodium iodide and iodine were all A.R. grade and all purchased from Sinopharm Chemical Reagent Co., Ltd, China. All reagents were used without further treating.

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 substrates for precipitating TiO_2 porous films. Sensitizing dye *cis*- $[(\text{dcbH}_2)_2\text{Ru}(\text{SCN})_2]$ and optical diffuser paste Ti-Nanoxide 300 were purchased from Solaronix SA.

2.2. Synthesis of PAA–PEG hybrid and preparation of polymer gel electrolyte

PAA–PEG hybrid was synthesized by the following processes. Firstly, 4.4 g PEG ($M_w = 1000$) was dissolved in 10 ml deionized water under stirring. The mixture was marked as A. Secondly, PAA was prepared by prepolymerization in water solution. 7.2 ml acrylic acid monomers were dissolved in 10 ml deionized water under stirring. Sodium pyrosulfite with an equivalent mol ratio of ammonium persulfate (2 wt% of monomers) were used as room temperature redox initiators to initiate polymerization reaction, the mixture was marked as B. When the temperature of mixture B rose quickly and higher than 80°C , the mixture A was dropped into the mixture B immediately under vigorous stirring to form homogeneous hybrid. The final hybrid was kept in the ambient environment for 1 h to finish the polymerization because of the exothermic reaction. Finally, the hybrid was kept in a vacuum oven at 80°C till the water in the hybrid was totally removed. Polymer gel electrolyte was prepared by soaking the hybrid in liquid electrolyte to absorbing saturation. The components of liquid electrolyte contain NaI, I_2 , Pyridine, NMP and GBL.

2.3. Assembling of quasi-solid-state dye-sensitized solar cell

The preparation of TiO_2 colloidal paste and double-layer film was according to the method reported by Wang et al. [23]. The TiO_2 colloidal paste was used to fabricate the transparent film

about $10 \mu\text{m}$. Then the optical diffuser film about $4 \mu\text{m}$ was printed above the transparent film by using Ti-Nanoxide 300 paste. After sintering at 500°C and cooling down to 80°C , the double-layer nanostructured TiO_2 electrodes were dye-coated by immersing them into a $2.5 \times 10^{-4} \text{ M}$ absolute ethanol solution of *cis*- $[(\text{dcbH}_2)_2\text{Ru}(\text{SCN})_2]$ for 24 h.

A quasi-solid-state dye-sensitized solar cell was fabricated by sandwiching a slice of polymer gel electrolyte between a dye-sensitized TiO_2 electrode and an electrodeposited platinum counter electrode [24]. The two electrodes were clipped together. Under the pressure of clips, polymer gel electrolyte was inserted into the pores of dye-sensitized TiO_2 nanostructure electrode to form good interfacial contact. A piece of cyanoacrylate adhesive ($15\text{--}30 \mu\text{m}$) was used as sealant. Bisphenol A epoxy resin (using ethylenediamine as epoxy hardener) was used for further sealing the cell.

2.4. Measurements

ATR-FTIR measurements were carried out using a Nicolet Impact 410 FTIR spectrometer. The liquid electrolyte absorbency (Q_{le}) of hybrid was defined as:

$$Q_{\text{le}} = \frac{W - W_0}{W} \times 100\% \quad (1)$$

where W is the weight of swollen hybrid, and W_0 is the original weight of dry hybrid. The Q_{le} values of samples were calculated according to Eq. (1). Ionic conductivity was measured by using model DDB-6200 digitized conductivity meter (Shanghai Reici Instrument Factory, China) at 30°C . The instrument was calibrated with 0.01 M KCl aqueous solution prior to experiments.

The photovoltaic tests of quasi-solid-state dye-sensitized solar cells were 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) under ambient atmosphere. The incident light intensity and the active cell area were 100 mW cm^{-2} and 0.2 cm^2 ($0.4 \text{ cm} \times 0.5 \text{ cm}$), respectively. The fill factor (FF) and overall energy conversion efficiency (η) of the cell were calculated by the following equations:

$$\text{FF} = \frac{V_{\text{max}} \times J_{\text{max}}}{V_{\text{OC}} \times J_{\text{SC}}} \quad (2)$$

$$\eta (\%) = \frac{V_{\text{max}} \times J_{\text{max}}}{P_{\text{in}}} \times 100\% = \frac{V_{\text{OC}} \times J_{\text{SC}} \times \text{FF}}{P_{\text{in}}} \times 100\% \quad (3)$$

where J_{SC} is the short-circuit current density (mA cm^{-2}), V_{OC} is the open-circuit voltage (V), P_{in} is the incident light power, and J_{max} (mA cm^{-2}) and V_{max} (V) are the current density and voltage in the J – V curves, respectively, at the point of maximum power output.

3. Results and discussion

The mixing of PAA and PEG can lead to the formation of stable hybrid based on hydrogen bonding between the hydroxyl

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