

Influence of solvent on the poly (acrylic acid)-oligo-(ethylene glycol) polymer gel electrolyte and the performance of quasi-solid-state dye-sensitized solar cells

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Abstract

The influence of solvents on the property of poly (acrylic acid)-oligo-(ethylene glycol) polymer gel electrolyte and photovoltaic performance of quasi-solid-state dye-sensitized solar cells (DSSCs) were investigated. Solvents or mixed solvents with large donor number enhance the liquid electrolyte absorbency, which further influences the ionic conductivity of polymer gel electrolyte. A polymer gel electrolyte with ionic conductivity of 4.45 mS cm^{-1} was obtained by using poly (acrylic acid)-oligo-(ethylene glycol) as polymer matrix, and absorbing 30 vol.% *N*-methyl pyrrolidone and 70 vol.% γ -butyrolactone with 0.5 M NaI and 0.05 M I_2 . By using this polymer gel electrolyte coupling with 0.4 M pyridine additive, a quasi-solid-state dye-sensitized solar cell with conversion efficiency of 4.74% was obtained under irradiation of 100 mW cm^{-2} (AM 1.5).

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1. Introduction

Dye-sensitized solar cells (DSSCs) are one of the promising candidates for the next generation of solar cells because of their simple structure with relatively high conversion efficiencies, inexpensive fabrication procedures in contrast with amorphous silicon solar cells [1–3]. DSSCs based on liquid electrolyte (marked as LE-DSSCs) have already achieved a record conversion efficiency of 11% [4], however, the poor long-term stability of liquid electrolyte obstructs their practical uses [5]. Thus, work has been done to replace liquid electrolyte with p-type semiconductors or hole-conducting materials but currently with moderate success [6,7]. Recently, the interest in quasi-solid-state DSSCs (marked as QS-DSSCs), especially prepared by using polymer gel electrolyte, has risen immensely [8–12]. The merits of polymer gel electrolyte include moderate stability, high ionic conductivity, excellent filling property between electrolyte layer

and TiO_2 film, which meet the requirements of QS-DSSCs. The components of polymer gel electrolyte for QS-DSSCs include polymer matrix, organic solvents, iodide-triiodide redox couples. As components of polymer gel electrolyte, organic solvents play an important role in photovoltaic performance of DSSCs, which was demonstrated by us and others [13–15].

Recently, we have reported a new kind of thermo-irreversible polymer gel electrolyte based on poly (acrylic acid)-poly (ethylene glycol) (PAA-PEG) hybrid for QS-DSSCs [16,17]. It is known that the high liquid electrolyte content in gel electrolyte is prerequisite for fabricating high efficient QS-DSSCs [18]. Due to polyacid character of PAA-PEG hybrid, it is feasible to obtain high liquid electrolyte absorbency of the hybrid by using Lewis basic solvents or mixed solvents in liquid electrolyte. According to the definition of Lewis basic, the solvents with high donor number (DN) show high Lewis basicity. The experimental results show that the PAA-PEG hybrid cannot swell in organic solvents with donor number lower than 15. So some conventional and popular solvents usually used in preparing DSSCs such as acetonitrile (DN: 14.1), methoxyacetonitrile (DN: 14.6) do not adapt to PAA-PEG

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hybrid [14,15]. While solvents with high basicity will affect the amount of absorbed dye on the surface of TiO₂ film or even fade the dye-coated TiO₂ film. Taking the above factors into account, γ -butyrolactone (GBL), *N*-methyl pyrrolidone (NMP), propylene carbonate (PC) solvents were selected as organic solvent components for PAA-PEG hybrid. The swelling of PAA-PEG hybrid mainly depends on Lewis acid-basic interaction between polymer matrix and organic solvents, the solvents or mixed solvents with different donor number will affect the liquid electrolyte absorbency of the hybrid, which determines the ionic conductivity of polymer gel electrolyte and further influence the photovoltaic performance of QS-DSSCs.

In this paper, the influence of organic solvents or mixed solvents with different donor number on the property of polymer gel electrolyte, and the photovoltaic performance of QS-DSSCs was investigated. The photovoltaic performance of the QS-DSSCs and the LE-DSSCs with the same organic solvent was compared. It is known [19] that the electric additives such as pyridine and 4-*tert*-butylpyridine are efficient matters for optimizing photovoltaic performance by increasing open-circuit voltage and fill factor, while little decreasing short-circuit current density, which causes the enhancement of light-to-electricity conversion efficiency of DSSCs. So the influence of pyridine additive on the photovoltaic performance of QS-DSSCs is also discussed in the paper.

2. Experimental

2.1. Materials

Poly (ethylene glycol) with average molecular weight 1000, acrylic acid monomers, ammonium persulfate, sodium pyrosulfite, titanium isopropoxide, nitric acid, glacial acetic acid, terpeneol, ethyl cellulose, ethanol, γ -butyrolactone (GBL), *N*-methylpyrrolidone (NMP), propylene carbonate (PC), 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 Ω cm⁻², purchased from Hartford Glass Co. USA) were used as substrates for precipitating TiO₂ porous films. Sensitizing dye *cis*-[(dcbH₂)₂Ru(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 hybrids were 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. A 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 moved into a vacuum oven at 80 °C for 24 h to remove the water in the hybrid. Polymer gel electrolyte was prepared by soaking the hybrid in liquid electrolyte till the liquid electrolyte absorbency of the hybrid obtaining a constant value. The latter was composed of 0.5 M NaI, 0.05 M I₂ in organic solvents or mixed organic solvents including GBL, NMP, PC and their mixed solvents. PY was used as electrolytic additive.

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

The preparation of TiO₂ colloidal paste and double-layer film was according to the method reported by Wang [20]. The TiO₂ colloidal paste was used to fabricate the transparent film about 10 μ m. Then the optical diffuser film about 4 μ 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 TiO₂ electrodes were dye-coated by immersing them into a 2.5×10^{-4} M absolute ethanol solution of *cis*-[(dcbH₂)₂Ru(SCN)₂] for 24 h.

A QS-DSSC was fabricated by sandwiching a slice of polymer gel electrolyte between a dye-sensitized TiO₂ electrode and an electrodeposited platinum counter electrode [21]. The two electrodes were clipped together and a piece of cyanoacrylate adhesive (15–30 μ m) was used as sealant. Bisphenol A epoxy resin (using ethylenediamine as epoxy hardener) was used for further sealing the cell.

2.4. Measurements

The liquid electrolyte absorbency (Q_{le}) of the hybrid was defined as:

$$Q_{le} = \frac{W - W_0}{W_0} \quad (1)$$

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). The instrument was calibrated with 0.01 M KCl aqueous solution prior to experiments. The donor numbers of the mixed solvents were estimated by the additivity rule, described by the following equation [14]:

$$D_{\text{mixed}} = \left(\frac{x}{100}\right) D_{\text{added}} + \left(\frac{100-x}{100}\right) D_{\text{GBL}} \quad (2)$$

where D_{mixed} , D_{added} and D_{GBL} are the donor numbers of the mixed solvent, the added solvent and GBL, respectively. x is the volume percentage of added solvent in the mixed solvent.

The photovoltaic tests of QS-DSSCs and LE-DSSCs were carried out by measuring the J - V character curves under irradiation of white light from a 100 W xenon arc lamp (XQ-500W,

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