



Efficient quasi-solid-state dye-sensitized solar cells employing polyaniline and polypyrrole incorporated microporous conducting gel electrolytes



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HIGHLIGHTS

- PANi or PPy is incorporated into 3D framework of microporous PAA–PEG matrix.
- The electrocatalytic reaction of triiodides is conducted into conducting gel electrolyte.
- Liquid electrolyte is driven by osmotic pressure and capillary diffusion.
- An power conversion efficiency of 7.12% is recorded.

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ABSTRACT

With an aim of elevating liquid electrolyte loading, ionic conductivity, and electrocatalytic activity of gel electrolyte toward triiodides, freeze-dried microporous poly(acrylic acid)–poly(ethylene glycol) (PAA–PEG) matrix is employed to uptake conducting polymers, such as polyaniline (PANi) and polypyrrole (PPy). An ionic conductivity of 19.18 mS cm⁻¹ at 65 °C is obtained from PAA–PEG/PANi conducting gel electrolyte which is nearly similar to 19.22 mS cm⁻¹ from pure liquid electrolyte. The conducting polymers can form interconnected channels within insulating microporous PAA–PEG matrix, therefore, the reduction reaction of triiodide ions in the dye-sensitized solar cells (DSSCs) can be extended from Pt/gel electrolyte interface to both interface and three-dimensional framework of microporous conducting gel electrolyte. The resultant DSSCs from PAA–PEG/PANi and PAA–PEG/PPy conducting gel electrolytes display power conversion efficiencies of 7.12% and 6.53%, respectively, which are much higher than 5.02% from pure PAA–PEG-based DSSC. The new concept along with easy fabrication promises the microporous conducting gel electrolytes to be good alternatives in efficient quasi-solid-state DSSCs.

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1. Introduction

As a promising solution to energy depletion, environmental pollution, and ecological destruction, dye-sensitized solar cells (DSSCs) have attracted growing interests because of their merits on easy fabrication, low cost, and relatively high power conversion efficiency [1–3]. A typical DSSC, composing of dye-sensitized TiO₂ anode, Pt counter electrode, and I⁻/I₃⁻ redox species, is an electrochemical device converting solar energy to electricity with no pollutant emission [4]. The leakage of liquid electrolyte and evaporation of organic solvent such as nitriles are always burdens for

commercial application of DSSCs [5,6]. By addressing these two issues, full-solid-state electrolytes are preferred candidates in highly stable DSSC devices [7,8]. However, the full-solid-state electrolytes cannot provide a relatively high carrier transport in inorganic electrolytes (hole diffusion) or polymer electrolytes (movement of chain segments), resulting in a decreased redox kinetics. Polymer gel electrolytes, especially the ones formed by imbibing liquid electrolyte into three-dimensional (3D) frameworks of amphiphilic gel matrices combine the rapid ionic transfer of liquid electrolytes and relatively high stability of full-solid-state electrolytes [9–13]. The imbibed liquid electrolyte can migrate within the interconnected microporous structure of gel matrix and cannot leak from the sealed framework during cell fabrication. However, there is still a remaining problem for these gel electrolyte-based DSSCs in Pt counter electrode/gel electrolyte

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interface. In comparison with Pt counter electrode/liquid electrolyte interface, the Pt counter electrode/gel electrolyte interface has an elevated resistance for the electrocatalytic reaction of triiodide ions with refluxed electrons because of the poor contact of polymer gel electrolyte and solid-state Pt counter electrode. In our previous research, we open a concept of extending the electrocatalytic reaction of triiodides from gel electrolyte/Pt counter electrode interface to both interface and 3D framework of conducting gel electrolyte, in which monomers of conducting polymers (such as aniline, pyrrole) are imbibed and in-situ polymerized in the matrices [14–18]. The power conversion efficiency has been significantly enhanced from ~5.5% to ~6.5%. These conducting gel electrolytes suffer from low liquid electrolyte loading and therefore ionic conductivity because the swelling of gel matrices in liquid electrolyte obeys Flory theory [19], in which the driving force of liquid electrolyte is osmotic pressure present across the gel matrices.

Here we report another new concept of employing microporous structure into poly(acrylic acid)–poly(ethylene glycol) (PAA–PEG) semi-interpenetrating (semi-IPN) gel matrix as a way to increase liquid electrolyte loading and therefore ionic conductivity and electrocatalytic activity. To the best of our knowledge, there are no reports on the employment of microporous conducting gel matrices in efficient DSSCs. The loading of liquid electrolyte by PAA–PEG is primarily driven by both osmotic pressure present across the gel matrix and capillary diffusion from micropores [20], resulting an enhanced liquid electrolyte loading. Elevated redox couple loading along with enhanced electrocatalytic activity is expected to provide increased photovoltaic performances of quasi-solid-state DSSCs.

2. Experimental

2.1. Synthesis of PAA–PEG matrices

The PAA–PEG matrices were synthesized according to an aqueous solution polymerization route. In detail, a solution mixture consisting of acrylic acid monomer (8 ml, AA, analytical reagent) and PEG (4.4 g, $M_w = 20,000$, analytical reagent) was made by agitating in deionized water (15 ml) in a water-bath at 80 °C. Under vigorous agitation, crosslinker *N,N'*-(methylene)bisacrylamide (NMBA, 0.008 g) and initiator ammonium persulfate (APS, 0.225 g) were subsequently added to the above mixture. With the proceeding of polymerization, the viscosity increased gradually. When the viscosity of the PAA–PEG reached around 180 mPa s⁻¹, the reagent was poured into a Petri dish and cooled to room temperature with the formation of an elastic transparent gel. The PAA–PEG membranes were then molded into ϕ 2.5 cm die. After rinsing with deionized water, the membranes were dried under vacuum at 50 °C for 24 h.

2.2. Preparation of PANi and PPy

0.592 ml of aniline was dissolved in 20 ml of 1 M HCl aqueous solution to obtain a homogeneous mixture. 20 ml of 0.125 M APS aqueous solution was dipped in the above mixture within 30 min. The polymerization reaction was carried out at 0 °C. After 3 h, the resultant reactant was rinsed by 1 M HCl aqueous solution, filtrated, and finally vacuum dried at 60 °C for 24 h.

1 ml of pyrrole monomer was dipped in an aqueous solution of FeCl₃ containing 7.788 g of FeCl₃·6H₂O and 58 ml of deionized water. Under vigorous agitation, the polymerization reaction was carried out at 5 °C for 24 h. After being rinsed by aqueous solution, filtrated, and finally vacuum dried at 60 °C for 24 h, the resultant PPy powders were obtained.

2.3. Preparation of conducting gel electrolytes

The microporous PAA–PEG matrices were prepared by immersing dried PAA–PEG matrices in deionized water for 72 h to reach their swelling equilibrium, and subsequently freeze-dried under vacuum at –60 °C for 72 h. After that the matrices were immersed in liquid electrolyte consisting of redox electrolyte and PANi or PPy for 5 days to reach absorption equilibrium. The contents of PANi and PPy in liquid electrolyte were 2.10 and 1.38 g L⁻¹, respectively. A redox electrolyte consisted of 100 mM of tetraethylammonium iodide, 100 mM of tetramethylammonium iodide, 100 mM of tetrabutylammonium iodide, 100 mM of NaI, 100 mM of KI, 100 mM of LiI, 50 mM of I₂, and 500 mM of 4-*tert*-butyl-pyridine in 50 ml acetonitrile.

2.4. Assembly of quasi-solid-state DSSCs

A layer of TiO₂ nanocrystal anode film with a thickness of 10 μ m and active area of 0.09 cm² was prepared by coating the TiO₂ colloid using a screen printing technique, followed by sintering in air at 450 °C for 30 min. Subsequently, the TiO₂ film was soaked in a 0.5 mM N719 [cis-di(thiocyanato)-*N,N'*-bis(2,2'-bipyridyl-4-carboxylic acid-4-tetrabutylammonium carboxylate)], purchased from Dyesol LTD, Australia] ethanol solution for 24 h to uptake N719 dye for the fabrication of dye-sensitized TiO₂ photoanode. The quasi-solid-state DSSCs from PAA–PEG/PANi, PAA–PEG/PPy, or PAA–PEG gel electrolyte at an imbibition equilibrium were fabricated by sandwiching a slice of gel electrolyte with a thickness of around 1 mm between dye-sensitized TiO₂ anode and a Pt counter electrode.

2.5. Photovoltaic measurements

The photocurrent–voltage (*J*–*V*) curves of the assembled quasi-solid-state DSSC were recorded on an electrochemical workstation (CHI600E) under irradiation of a simulated solar light from a 100 W xenon arc lamp in ambient atmosphere. The incident light intensity was calibrated using a FZ-A type radiometer from Beijing Normal University Photoelectric Instrument Factory to control it at 100 mW cm⁻² (AM 1.5). Each DSSC device was measured five times to eliminate experimental error and a compromise *J*–*V* curve was employed.

2.6. Characterizations

The morphologies of the gel electrolytes were captured with a Zeiss Ultra plus field emission scanning electron microscopy (FESEM). To observe the internal 3D microstructure, swollen gel electrolytes were first cut into ultrathin film, followed by the loading into a chamber under freezing temperature and high vacuum to remove solvent. Fourier transform infrared spectrometry (FTIR) spectra were recorded on a Vertex 70 FTIR spectrometer (Bruker). The ionic conductivity of gel electrolyte was measured by using a pocket conductivity meter (DSSJ-308A, LeiCi Instruments). The instrument was calibrated with 0.01 M KCl aqueous solution prior to experiments. Tafel polarization curves of the symmetrical cells fabricated of samples were measured by CHI660E electrochemical workstation. The symmetrical dummy cells fabricated with two identical Pt electrodes (Pt electrode/gel electrolyte/Pt electrode). The electrochemical impedance spectroscopy (EIS) was carried out using a CHI660E electrochemical workstation at a constant temperature of 20 °C with an ac signal amplitude of 20 mV in the frequency range from 0.1 to 10⁵ Hz at 0 V dc bias in the dark. The cyclic voltammetry (CV) was also carried out on a CHI660E electrochemical workstation in a N₂-purged liquid electrolyte. The

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