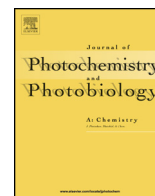




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Quasi-solid-state dye-sensitized solar cell based on gel electrolyte with high gel to solution transition temperature using low molecular mass organogelator



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ABSTRACT

Quasi-solid-state dye-sensitized solar cell (QS-DSSC) were fabricated using *N,N'*-1,4-butylenediybis-dodecanamide as low molecular mass organogelator (LMOG). The gel to solution transition temperature (T_{gel}) of this gel electrolyte is 115.8 °C which ensures a good intrinsic stability of the gel electrolyte at the QS-DSSC operating temperature. The kinetic processes of electron transport and recombination of DSSCs influenced by the gel electrolyte are investigated in detail. Due to the diffusion of redox species in the gel electrolyte is hindered by the crosslinked network and the electron recombination at the interface of the TiO_2 photoanode/electrolyte is accelerated, the incidental photon-to-electron conversion efficiency (IPCE) and the short circuit photocurrent density (J_{sc}) of the QS-DSSC are slightly decreased compared with those of liquid electrolyte based DSSC (L-DSSC). Remarkably, the QS-DSSC exhibits excellent thermal and light-soaking stabilities during accelerated aging tests for 1000 h. The QS-DSSC retains 90% of its initial photoelectric conversion efficiency, whereas that of L-DSSC only retains 83% under successive one sun light soaking with UV cut-off filter at 50 °C for 1000 h.

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1. Introduction

Dye-sensitized solar cells (DSSCs) have been regarded as a promising candidate for next generation solar energy conversion technology and attracted widespread attention over the past decade owing to their low cost, low energy consumption, environmentally friendly materials, simple fabrication process and comparatively high power conversion efficiency [1–3]. The electrolyte is one of key factors that affect the photovoltaic performance of DSSCs. High efficiencies have been achieved with the use of liquid electrolytes [2,4]. The conventional electrolyte

solvents used in DSSC are some organic solvents such as γ -butyrolactone [5–7], acetonitrile [8–10] and 3-methoxypropionitrile [11,12]. However, the volatile nature of organic solvents in traditional liquid electrolytes makes the sealing difficult, leading to stability problems which restrict the application and commercialization of DSSCs. As a result, many alternatives have been made to overcome this problem by replacing the liquid electrolytes, including the solid-state electrolytes such as the hole-conductors and polymer electrolytes [13–16]. However, derivative problems such as contact between hole transporting materials and the nanoporous semiconductor films are remain to be solved [17,18]. Therefore, quasi-solid-state electrolytes have attracted much attention for their good performance and stability which can be prepared by solidifying the liquid electrolytes using low molecular mass organogelators (LMOGs) [19,20], polymer gelators [21,22] or nanoparticles [23,24]. Compared with the two kinds of electrolyte, the gel electrolytes formed from the LMOGs exhibit three dimensional networks and possess higher ionic conductivity, good

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pore filling and penetration of the mesoporous TiO₂ film [25,26]. Importantly, the devices assembled by LMOGs exhibit good long-term stability because of high gel to solution transition temperature of LMOGs [27,28].

In this work, an amide derivative, *N,N'*-1,4-butylenediylbis-dodecanamide was synthesized and used as LMOG to fabricate a novel gel electrolyte with high T_{gel} . The charge transport mechanism of this gel electrolyte, kinetic processes of electron transport and recombination, as well as photovoltaic performance and stabilities of the corresponding QS-DSSC were investigated in detail.

2. Experimental

2.1. Preparation of the liquid and gel electrolytes

The *N,N'*-1,4-butylenediylbis-dodecanamide and the 1,2-dimethyl-3-propylimidazolium iodide (DMPII) were synthesized as previously reported [29,30]. The liquid electrolyte was composed of 0.9 mol L⁻¹ DMPII, 0.1 mol L⁻¹ anhydrous lithium iodide (LiI: 99%, Aldrich), 0.1 mol L⁻¹ iodine (I₂: 99%, Aldrich) and 0.5 mol L⁻¹ *N*-methylbenzimidazole (NMBI: 99%, Aldrich) in methoxypyridine (MePN). Moreover, the liquid electrolyte should divide into two parts, one part was for the reference electrolyte and applied to DSSC, and the other part was used for preparing the gel electrolyte. In addition, the gel electrolyte was formed by adding 8 wt% (vs. liquid electrolyte) *N,N'*-1,4-butylenediylbis-dodecanamide into the liquid electrolyte and heated to 120 °C under stirring until the gelator was dissolved. After cooling to room temperature, the gel electrolyte was obtained and used in QS-DSSC.

2.2. Fabrication of dye-sensitized solar cells

The colloidal TiO₂ microspheres were prepared by hydrolysis of titanium tetraisopropoxide as shown in the previous literature [31]. Nanocrystalline electrodes about 18 μm thick were obtained by screen-printing TiO₂ paste on FTO glass (TEC-8, LOF). After sintering at 450 °C for 30 min in air then cooling to about 120 °C, the nanoporous TiO₂ photoelectrode was immersed in an ethanol solution of 0.5 mmol L⁻¹ *cis*-dithiocyanate-*N,N*-bis-(4-carboxylate-4-tetrabutyl ammoniumcarboxylate-2,2-bipyridine) ruthenium(II) (N719 dye) for 14 h. The platinized counter electrodes were obtained by spraying H₂PtCl₆ solution to FTO glass followed by heating at 410 °C for 20 min. Then, the dyed nanoporous TiO₂ photoelectrode and the counter electrode were sealed together with a thermal adhesive film (Surlyn 1702, Dupont, USA). The liquid electrolyte was injected into the internal space between two electrodes through the hole on the counter electrode, which was later sealed by a cover glass and thermal adhesive film. In this way, the reference liquid electrolyte based DSSC was formed. The gel electrolyte was heated to 120 °C under stirring until the gel transforms into a liquid (hot solution) completely. Then, the gel electrolyte (hot liquid solution) was rapidly injected into the cell and the cell was sealed by a cover glass and thermal adhesive film. After cooling to room temperature, a uniform motionless gel layer was formed in the cell. According to this way, the gel electrolyte based QS-DSSC was prepared.

2.3. Differential scanning calorimetry (DSC)

The gel to solution transition temperature (T_{gel}) of the gel electrolyte was determined by differential scanning calorimeter (DSC-Q2000, TA, USA). Approximately 5–7 mg of the sample was weighed, sealed in an aluminum pan and heated at a rate of 10 °C min⁻¹ under nitrogen flow from 25 °C to 140 °C for DSC measurement.

2.4. Linear sweep voltammetry measurements

Linear sweep voltammograms were recorded on an electrochemical workstation (Autolab 320, Metrohm, Switzerland) at 25 °C in a two-electrode system equipped with a 1 mm radius platinum disk electrode (CHI102, CH Instruments Inc., USA) as the counter electrode and a reference electrode, a 5.0 μm radius platinum ultramicroelectrode (CHI107, CH Instruments Inc., USA) as the working electrode. The linear sweep voltammograms were obtained at a scan rate of 5 mV s⁻¹.

2.5. Electrochemical impedance spectroscopy (EIS) measurement

EIS measurements of DSSCs were recorded with an electrochemical workstation (Autolab 320, Metrohm, Switzerland). To measure the impedance, a direct-current bias at 690 mV, and perturbation amplitude of 10 mV within the frequency range from 10 mHz to 1 MHz was applied in darkness. The obtained impedance spectra were fitted with Z-view software (v2.8b, Scribner Associates, USA) in terms of a transmission line equivalent circuit model to interpret the characteristics of DSSC.

2.6. Controlled intensity modulated photocurrent/photovoltage spectroscopy (IMPS/IMVS) measurements

Controlled intensity modulated photocurrent/photovoltage spectroscopy (IMPS/IMVS) measurements is measured by an electrochemical workstation (IM6e, Zahner, Germany) with light emitting diodes (LED) (λ=610 nm) driven by a source supply (XPOT, Zahner, Germany).

2.7. Characterization of incidental photon-to-electron conversion efficiency (IPCE)

The photocurrent action spectra were recorded on a QE/IPCE measurement. The white light was emitted by a 300 W xenon lamp (69911, Newport, USA) and transported to a monochromatic light (400 ~ 800 nm) by a 1/4 m monochromator (74125 Oriel Cornerstone 260, Newport, USA), then the light beat down on the calibrated UV silicon photodetector (71675, Newport, USA) and the DSSC in turn, the monochromatic light intensity and the photocurrent for DSSC were measured by a dual channel power meter (2931-C, Newport, USA) and the calibrated UV silicon photodetector. In addition, the sampling wavelength interval is 10 nm and sampling time is 2 s.

2.8. Photovoltaic characterizations and stability tests

The incident light intensity was calibrated with a standard crystalline silicon solar cell before each experiment. The photovoltaic performance of DSSCs has an active area of 0.16 cm². With a filter (AM 1.5, 100 mW cm⁻²), The photovoltaic performance of DSSCs were measured by a Keithley 2420 digital source meter (Keithley, USA) and controlled by Test point software under a 450 W xenon lamp (Oriel, USA).

DSSCs covered with a UV cut-off filter (up to 394 nm) were irradiated at the open circuit under one sun light soaking (XQ3000, 100 mW cm⁻², Shanghai B.R. Science Instrument Co., Ltd, China) and the ambient temperature was set to 50 °C during the light soaking experiment. *J*-*V* measurements were carried out at room temperature after allowing these cells to cool down and equilibrate for 30 min. The *J*-*V* measurements were carried out from short circuit to open circuit with sampling delay time ranging from 1 ms to 1 s. The measuring integration time and step source level were fixed at 50 ms and 10 mV, respectively.

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