



Effect of alkyl chain length of imidazolium cations on the electron transport and recombination kinetics in ionic gel electrolytes based quasi-solid-state dye-sensitized solar cells



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ABSTRACT

A series of stable quasi-solid-state dye-sensitized solar cells (QS-DSSCs) are prepared by the 12-hydroxystearic acid as low molecular mass organogelator (LMOG) to gelate the ionic liquid with different alkyl chain lengths (3, 4, and 7). The influence of alkyl chain length of imidazolium cations (Im^+) on the kinetic processes of electron transport and recombination are investigated by Electrochemical Impedance Spectroscopy (EIS) and intensity-modulated photocurrent spectroscopy/intensity-modulated photovoltage spectroscopy (IMPS/IMVS). It is found that the ionic gel electrolytes (IGEs) with different alkyl chain lengths of Im^+ can influence the competitive adsorption effects of imidazolium cations (Im^+) and Li^+ , and further affect the charge diffusion, the electron recombination/transport processes, the shift of TiO_2 conduction band edge and surface states distribution. The IGE with longer alkyl chain length of Im^+ can prolong the electron recombination lifetime, promote the incidental photon-to-electron conversion efficiency (IPCE) and the short circuit photocurrent density (J_{sc}). An excellent QS-DSSC based on the IGE with the longer alkyl chain of Im^+ gives the highest photoelectric conversion efficiency. Moreover, all the QS-DSSCs based on IGEs exhibit excellent durability without losing their photovoltaic performances during the accelerated thermal and light-soaking test. These results are very important to the researches on the electrochemical mechanism and application of QS-DSSCs based on IGEs.

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1. Introduction

Dye-sensitized solar cells (DSSCs) are the attractive candidates for the next generation of solar energy conversion technologies due to their low-cost materials, simple fabrication process and high energy conversion efficiencies [1,2]. Electrolytes, as the important components in DSSCs, play an important role in the photovoltaic performances of DSSCs. The photoelectric conversion efficiency of DSSCs based on liquid electrolyte has achieved 13% [3], however, the leaking and sealing problem of DSSCs which result from the organic solvent are a challenge for the application and

commercialization of DSSCs. Room-temperature ionic liquids (ILs) have received intense interest in recent years and used as excellent solvents for DSSCs due to their desirable properties such as negligible volatility, high electrochemical and thermal stability, and inherent ionic conductivity [4,5]. The DSSCs based on ILs can relieve the leaking problem to a certain degree, but in order to further improve the stability of DSSCs, low molecular mass organogelators (LMOGs) are used to gelate ILs to obtain the ionic gel electrolytes (IGEs) based quasi-solid-state DSSCs (QS-DSSCs) and have aroused wide concern. So far, imidazolium cation (Im^+) based ILs have been widely applied in QS-DSSCs. Nevertheless, how chemical structures of Im^+ influence the kinetic processes of electron transport and recombination and the photovoltaic performance of the QS-DSSCs remains relatively unexplored.

It is well known that cations which exist in the electrolyte such as Li^+ and Im^+ can affect the quantity and distribution of surface states, the rate of charge diffusion and the kinetics of interfacial

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electron transfer [6,7]. A longer cation side chain is commonly accompanied by lower density, slower diffusion, and higher viscosity [8,9]. Moreover, due to the electrostatic effect, Im^+ can adsorb onto the TiO_2 surfaces to form Im^+ multilayer and further influence the adsorption and function of Li^+ . Many reports had been published about the photovoltaic performances of the QS-DSSCs based on IGEs [10–13], but there is little research on the kinetic response with the change of the alkyl chain length of Im^+ in IGEs based QS-DSSCs.

In this work, we introduced 12-hydroxystearic acid as a LMOG to gelate the 1-Methyl-3-propylimidazolium iodide (MPII), 1-Methyl-3-butylimidazolium iodide (BMII), 1-Methyl-3-hexylimidazolium iodide (HMII) and obtained the IGEs based QS-DSSCs. To investigate the differences in kinetic processes of electron transport and recombination in these QS-DSSCs, intensity-modulated photocurrent spectroscopy/intensity-modulated photovoltage spectroscopy (IMPS/IMVS) measurements, Electrochemical impedance spectroscopy (EIS) measurement and equivalent circuit analysis were carried out. This work is important and necessary to further understand the detailed mechanism of the influence of the different IGEs with different alkyl chain lengths of Im^+ on the QS-DSSCs performances.

2. Experiment detail

2.1. Electrolyte preparation

1-Methyl-3-propylimidazolium iodide (MPII), 1-Methyl-3-butylimidazolium iodide (BMII) and 1-Methyl-3-hexylimidazolium iodide (HMII) were prepared as reported previously [14,15]. The ionic liquid electrolytes (ILEs) for QS-DSSC were composed of 0.35 mol L^{-1} iodine (I_2 : 99%, Aldrich), 0.02 mol L^{-1} anhydrous lithium iodide (LiI: 99%, Aldrich) and 0.5 mol L^{-1} N-methylbenzimidazole (NMBI: 99%, Aldrich) in MPII, BMII and HMII, respectively. IGEs were prepared by adding 20 wt% (vs. ILE) 12-hydroxystearic acid (99%, Aldrich) into ILEs and heated under stirring until the gelators melted. After cooling to room temperature, the IGEs were formed. The IGEs based on MPII, MBII and HMII correspond to IGE P, IGE B and IGE H, respectively.

2.2. Fabrication of dye-sensitized solar cells

The TiO_2 nanocrystalline electrodes about $11.3 \mu\text{m}$ thickness were obtained by screen-printing TiO_2 paste on FTO glass (TEC-8, LOF). After sintering at 450°C for 30 min in air then cooling to 120°C , the nanoporous TiO_2 photoelectrodes were immersed in an ethanol solution of 0.5 mmol L^{-1} cis-dithiocyanate-N,N-bis-(4-carboxylate-4-tetrabutylammoniumcarboxylate-2,2-bipyridine) ruthenium(II) (N719 dye) for 14 h. The platinized counter electrodes were obtained by spraying H_2PtCl_6 solution to FTO glass followed by heating at 410°C for 20 min. DSSCs were assembled by sealing the dyed nanoporous TiO_2 photoelectrode and the counter electrode with a thermal adhesive film (Surlyn 1702, Dupont, USA). The ILEs were injected into the internal space between of two electrodes through the hole on the counter electrode, which was later sealed by a cover glass and thermal adhesive film. The IGEs were heated to 135°C under stirring until the gel transform to liquid completely. Then, the electrolytes (hot solutions) were rapidly injected into the cells respectively and the cells were sealed as the same as the ILEs based DSSCs. After cooling to room temperature, a uniform motionless gel layer was formed in cell.

2.3. Linear sweep voltammetry measurements

Linear sweep voltammograms were recorded on an electrochemical workstation (Autolab 320, Metrohm, Switzerland) at

25°C in two-electrode system equipped with a $5.0 \mu\text{m}$ platinum ultramicroelectrode (CHI107, CH Instruments Inc. USA) as working electrode, a 1 mm radius platinum disk electrode (CHI102, CH Instruments Inc. USA) as counter electrode and reference electrode [16,17]. The linear sweep voltammograms were obtained at scan rate of 5 mV s^{-1} .

2.4. Differential scanning calorimetry

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 each sample was weighed and sealed in an aluminum pan and heated at a rate of $10^\circ\text{C min}^{-1}$ under nitrogen flow for DSC measurement.

2.5. Electrochemical impedance spectroscopy (EIS) measurement

EIS measurement of DSSCs was recorded with an electrochemical analyzer (Autolab 320, Metrohm, Switzerland). To measure the impedance, a direct-current bias at -630 mV , and a perturbation amplitude of 10 mV within the frequency range from 1 MHz to 10 mHz was applied in dark. The obtained impedance spectra were fitted by Z-view software (v2.8b, Scribner Associates, USA) in terms of a transmission line equivalent circuit model to interpret the characteristics of DSSCs [18–21].

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

The experimental setup for IMPS and IMVS measurements have been described elsewhere [22,23]. Intensity-modulated measurements were carried out by an electrochemical workstation (IM6e, Zahner, Germany) with light emitting diodes (LED) ($\lambda = 610 \text{ nm}$) driven by a source supply (XPOT, Zahner, Germany). The LED provided both the dc and ac components of the illumination. A small ac component is 10% or less than that of the dc component and the frequency range was 3 kHz to 300 mHz .

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

The photocurrent action spectra were recorded on a QE/IPCE measurement kit consisting of a 300 W xenon lamp (69911, Newport, USA), a $1/4 \text{ m}$ monochromator (74125 Oriol Cornerstone 260, Newport, USA), a dual channel power meter (2931-C, Newport, USA) and the calibrated UV silicon photodetector (71675, Newport, USA).

2.8. Photovoltaic characterizations and stability tests

The photovoltaic performance of DSSCs with the active area of 0.16 cm^2 with black mask were measured by a Keithley 2420 digital source meter (Keithley, USA), and controlled by Test point software under a 450 W xenon lamp (Oriol, USA) with a filter (AM 1.5, 100 mW cm^{-2}). The incident light intensity was calibrated with a standard crystalline silicon solar cell before each experiment.

Hermetically sealed cells were used for long-term stability tests. The cells were stored in the oven at 60°C for thermal stress experiment. Furthermore, the successive one sun light soaking experiment was also carried out. DSSCs covered with a UV cutoff filter (up to 394 nm) were irradiated at open circuit under AM 1.5 (XQ3000 , 100 mW cm^{-2} , Shanghai B.R. Science Instrument Co., Ltd China) and the air 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.

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