



Full-ionic liquid gel electrolytes: Enhanced photovoltaic performances in dye-sensitized solar cells



Qinghua Li^a, Qunwei Tang^{b,*}, Benlin He^b, Peizhi Yang^c

^aKey Laboratory of Nondestructive, Ministry of Education, Nanchang Hangkong University, Nanchang 330063, Jiangxi Province, PR China

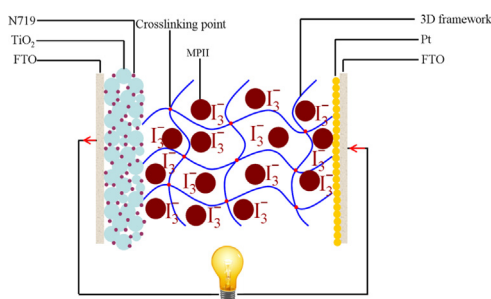
^bInstitute of Materials Science and Engineering, Ocean University of China, Qingdao 266100, Shandong Province, PR China

^cKey Laboratory of Advanced Technique & Preparation for Renewable Energy Materials, Ministry of Education, Yunnan Normal University, Kunming 650092, PR China

HIGHLIGHTS

- Ionic liquid-imbibed gel electrolyte is synthesized to replace traditional liquid electrolyte.
- Nonvolatility of ionic liquid is expected to enhance the long-term stability of DSSCs.
- The ionic conductivity of ionic liquid-imbibed gel electrolyte is enhanced.
- The conversion efficiency of the quasi-solid-state DSSC is 7.19%.

GRAPHICAL ABSTRACT



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ABSTRACT

Liquid electrolytes containing redox species have been widely used in dye-sensitized solar cells (DSSCs), whereas the volatility of organic solvents has been a tremendous obstacle for their commercial application. To assemble durable DSSCs, here we report the synthesis of full-ionic liquid electrolyte, in which 1-butyl-3-methylimidazolium nitrate is employed as solvent and 1-methyl-3-propylimidazolium iodide is iodide source. Using the imbibition performance of amphiphilic poly(acrylic acid/gelatin) [poly(AA/GR)] and poly(acrylic acid/cetyltrimethyl ammonium bromide) [poly(AA/CTAB)] matrices, full-ionic liquid electrolytes are imbibed into three-dimensional framework of poly(AA/GR) or poly(AA/CTAB) to form stable gel electrolytes. Room-temperature ionic conductivities as high as 17.82 and 18.44 mS cm⁻¹ are recorded from full-ionic liquid imbibed poly(AA/GR) and poly(AA/CTAB) gel electrolytes, respectively. Promising power conversion efficiencies of 7.19% and 7.15% are determined from their DSSC devices in comparison with 6.55% and 6.12% from traditional acetonitrile-based poly(AA/GR) and poly(AA/CTAB) gel electrolytes, respectively. The new concept along with easy fabrication demonstrates the full-ionic liquid electrolytes to be good alternatives for robust gel electrolytes in quasi-solid-state DSSCs.

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1. Introduction

Dye-sensitized solar cell (DSSC), an electrochemical device directly converting solar energy to electricity, has attracted growing

interests because of their merits on relatively high power conversion efficiency, low fabrication cost, and environmental-friendliness [1–6]. A typical DSSC device is composed of a dye sensitized TiO₂ photoanode, a counter electrode, and redox electrolyte. The task of redox electrolyte is to regenerate excited dye molecules and to be subsequently reduced by the counter electrode. To obtain higher reaction kinetics, organic solvents such as

* Corresponding author. Tel./fax: +86 532 66781690.

E-mail address: tangqunwei@ouc.edu.cn (Q. Tang).

acetonitrile are always employed as mediums for the transportation of iodide/triiodide (I^-/I_3^-) redox couples [7–10]. However, low ionic conductivity, insufficient penetration of the electrolyte into nanoporous structure of TiO_2 film, and volatility of organic solvents have been tremendous obstacles for the commercial application of DSSCs [11–13]. Therefore, it is a prerequisite to find an alternative solvent with nonvolatility, easy synthesis, low cost and environmental-friendly natures for I^-/I_3^- loading. By addressing these issues, room-temperature ionic liquids are preferred candidates.

Room-temperature ionic liquids, liquid state within temperature of 100 °C, are defined as ionic solvents composed of positively and negatively charged ions. They are always characterized with high glass transition temperature [14], reasonable thermal stability [15], high conductivity [16], and especially good dissolution to iodine and iodide [17]. Up to now, there are few reports on the employment of full-ionic liquid electrolyte in DSSCs, not to mention full-ionic liquid electrolyte imbibed three-dimensional (3D) amphiphilic framework-based quasi-solid-state DSSCs.

In the current work, 3D poly(acrylic acid/glycerol) [poly(AA/GR)] and poly(acrylic acid/cetyltrimethyl ammonium bromide) [poly(AA/CTAB)] polymer composites are employed as placeholders for full-ionic liquid loading to replace acetonitrile-contained iodide electrolytes [18,19]. Crosslinked poly(acrylic acid) is a typical 3D framework capable of absorbing enormous aqueous solution because of decoration with hydrophilic $-COOH$ groups, however, modification of poly(acrylic acid) by glycerol (GR) or cetyltrimethyl ammonium bromide (CTAB) can generate amphiphilicity to poly(AA/GR) or poly(AA/CTAB). The imbibed full-ionic liquid electrolyte in 3D framework of poly(AA/GR) or poly(AA/CTAB) can not leak even during cell assembly and operation. The resultant gel electrolytes are subsequently assembled into quasi-solid-state DSSC devices, which are expected to be honored by long-term stability. The objectives of this study are the synthesis of full-ionic liquid imbibed poly(AA/GR) and poly(AA/CTAB) gel electrolytes and their characterizations as well as the photovoltaic performances of assembled DSSCs from the gel electrolytes.

2. Experimental

2.1. Materials

Unless noted otherwise, chemicals were purchased from Sigma–Aldrich and used as received.

2.2. Synthesis of 3D poly(AA/GR) matrix

Poly(AA/GR) matrix was synthesized by the procedures: 5 ml of GR and 10 g of acrylic acid (AA) were dispersed in 10 ml of deionized water. Subsequently, initiator potassium peroxydisulfate (KPS) (mass ratio of KPS to AA was 0.008) and crosslinker *N,N'*-methylene bisacrylamide (NMBA) (mass ratio of NMBA to AA was 0.0005) were added to the mixed solution. When the viscosity of the poly(AA/GR) prepolymers reached around 180 mPa s⁻¹, the reagent was poured into a petri dish and cooled to room temperature until the formation of an elastic gel. After rinsing with excess deionized water, the samples were vacuum dried at 80 °C. Finally, the sample was vacuum dried at 80 °C for more than 12 h.

2.3. Synthesis of 3D poly(AA/CTAB) matrix

Poly(AA/CTAB) matrix was synthesized by the procedures: 1 g of cetyltrimethyl ammonium bromide (CTAB) and 10 g of AA were dispersed in 10 ml of deionized water. Subsequently, initiator KPS (mass ratio of KPS to AA was 0.008) and crosslinker NMBA (mass

ratio of NMBA to AA was 0.0005) were added to the mixed solution. When the viscosity of the poly(AA/CTAB) prepolymers reached around 180 mPa s⁻¹, the reagent was poured into a petri dish and cooled to room temperature until the formation of an elastic gel. After rinsing with excess deionized water, the samples were vacuum dried at 80 °C. Finally, the sample was vacuum dried at 80 °C for more than 12 h.

2.4. Synthesis of [AMIM]NO₃, [AMIM]BF₄, and MPII

The [AMIM]NO₃, [AMIM]BF₄, and 1-methyl-3-propylimidazolium iodide (MPII) were synthesized by following steps [20,21]: A mixture of 23.0 g of *N*-methyl-imidazolium and 25.3 g of allyl nitrate was vigorously agitated in a three-necked flask equipped with mechanical stirrer and thermometer. The excess allyl nitrate was removed by keeping temperature at 60–65 °C for 6–7 h, while the surplus *N*-methyl-imidazolium was extracted by vacuum drying at 80 °C for 48 h to obtain a transparently viscous liquid. 35.0 g of the as-synthesized viscous liquid, 20.0 g of silver nitrate, and 50.0 ml of methanol were mixed in another three-necked flask at 40 °C for 48 h to obtain [AMIM]NO₃. The unreacted sodium bicarbonate and methanol were removed in filtering and evaporating processes.

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Methyl imidazole and iodopropane with a molar ratio of 1: 1.1 were mixed in a 250 ml neck boiling flask which was sealed by toluene. The mixture was deaerated by bubbling N₂ for 5 min and dark-agitated at 35 °C for 24 h. Resultant product was thoroughly rinsed with anhydrous ether and shaken for 20 min and then kept stilling for 1 h until the liquid became brownish red. After rotary evaporation at 30 °C for 3 h, final MPII can be obtained.

2.5. Fabrication of full-ionic liquid imbibed gel electrolytes

Full-ionic liquid imbibed poly(AA/GR) gel electrolyte was prepared according to the following procedures [22–25]: 0.2 g of poly(AA/GR) was immersed in an ionic liquid electrolyte solution at ambient temperature, resulting in the diffusion of full-ionic liquid electrolyte into poly(AA/GR) framework and formation of a swollen sample to reach absorption saturation. The ionic liquid electrolyte consisted of 0.5 M I₂, 0.01 M LiI in mixed organic solvent of 40 vol% MPII, 50 vol% [AMIM]NO₃, and 10 vol% *N*-methyl pyrrolidone. Ionic liquid imbibed poly(AA/CTAB) gel electrolyte was prepared according to the following procedures: 0.2 g of poly(AA/CTAB) was immersed in an IL electrolyte solution at ambient temperature, resulting in the diffusion of RTIL electrolyte into the poly(AA/CTAB) network and formation of a swollen sample to reach absorption saturation. The RTIL electrolyte consisted of 0.5 M I₂, 0.01 M LiI in mixed organic solvent of 40 vol% MPII, 50 vol% [AMIM]BF₄, and 10 vol% *N*-methyl pyrrolidone.

The ionic liquid loading could be calculated according to the following equation:

$$\text{Ionic liquid loading (g g}^{-1}\text{)} = \frac{W_t - W_d}{W_d} \quad (1)$$

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