



Research paper

Dye-sensitized solar cells using ionic liquids as redox mediator

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ABSTRACT

In this research, the influence of ionic liquid on the conversion efficiency, incident photons to converted electrons (IPCE) and performance of fabricated solar cell was investigated using various ionic liquids. Ionic liquids with different substituents and ions were prepared and used as redox mediators in dye-sensitized solar cells (DSSCs). Ionic liquids were characterized ¹H and ¹³C NMR spectra. We practically investigated the performance of ionic liquid salts were used as the mobile ions and found that the efficiencies of DSSCs were increased up to 40% comparing commercial electrolyte system. The ionic liquid compounds were incorporated in DSSCs to obtain an efficient charge transfer, solving the corrosion problem of platinum layer in counter electrode compared to commercial electrolyte.

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1. Introduction

Solar cells based dye-sensitized have attracted much attention since a 1991 research breakthrough. These type solar cells are memorable for attaining high solar power conversion with easy procedure and low costs of manufacturing [1]. In the literature, different strategies have been suggested to enhance the performance of DSSCs such as photoanode [2], dyes [3], counter electrodes [4] and others [5]. Ionic liquids (IL) is composed of an organic as a salt in the liquid state whose melting point is below 100 °C. Ionic liquids are described as having many potential applications. They are powerful solvents and electrically conducting fluids (electrolytes). Ionic liquid have attention to study their fabrication characteristics, such as high heat capacity, excellent ionic conductivity, outstanding solubility, superior chemical and thermal and stability, strong polarity [6–12].

Nevertheless, the problems of corrosion and conductivity thus shortage of stability limit the commercial DSSCs [13]. Moreover, commercial electrolytes were published to tend fast recombination reactions, thus resulting low performance. To settle these troubles and enhance the stability of DSSCs, ionic liquid-based DSSCs have

been progressing. Chen et al. prepared an ionic liquid and synthesis the electrolyte to enhance stability of DSSCs [12].

One of the key parameter of DSSC is the selection of electrolyte which provides the internal electrical conductivity. The use of liquid electrolytes demands perfect sealing of the device to avoid leakage and evaporation of the solvents. Also, current electrolytes suffer from poor stability due to a decrease of tri-iodide concentration through the sublimation of iodine. Both of these issues affect the long term stability and the performance of the liquid electrolyte.

In this paper, various ionic liquids compounds samples were synthesized and characterized in our laboratory. All samples were applied in fabricated DSSCs as redox mediator in this work. The aim of this study is to investigate systematically the photovoltaic performances according to the effect of side chains in imidazolium salts. The commercial iodide based redox electrolyte was used for comparison. Photovoltaic parameters and the incident photons to converted electrons (IPCE) of the DSSCs were studied. The synthesized butyl substituted imidazolium iodine salt showed a power conversion efficiency of 5.17% under the simulated air mass 1.5 solar spectrum illumination at 100 mW cm⁻².

2. Experimental

2.1. General considerations

All reagents were purchased from commercial sources and used as received. 1-substituted imidazoles were synthesized according to the literature [14]. ¹H NMR (400 MHz) and ¹³C NMR

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(100 MHz) spectra were measured on Varian AS 400 Mercury spectrometers and tetramethylsilane (TMS) was used as the internal standard. All coupling constants (J values) were reported in Hertz (Hz). Elemental analyses were performed on a Perkin-Elmer PE 2400 elemental analyzer.

2.2. General procedures of ionic liquids

The solution of imidazole (1.0 mmol) and alkyl halide (1.0 mmol) in toluene (1 mL) was heated overnight. The solvent was decanted. The residue was washed with Et_2O (3×5 mL) and then dried under reduced pressure. Molecular structures were presented in Fig. 1.

2.3. Fabrication of DSSCs

Photoanode was fabricated using high transparency TiO_2 nanoparticles (particle size of 20 nm) pastes purchased from Dyesol. A conductive glass sheet fluorine-doped tin oxide FTO substrates of $2 \text{ cm} \times 1.5 \text{ cm}$, purchased from Dyesol (TEC8 Glass Plates, 8 ohm/square) was first cleaned in distilled water, and finally with acetone using an ultrasonic bath. The TiO_2 paste as mesoporous layer was coated onto FTO glass, using doctor-blade method between two parallel adhesive Scotch tapes. After drying, this paste was gradually sintered at or 500°C for 30 min, this results in a transparent sintered layer, with a film thickness of approximately 10–12 μm . The film thickness was controlled by scotch tape. Then, the TiO_2 paste was immersed into a 0.5 mM of N719 dye solution (N719 Industry Standard Dye-Dyesol Company) and kept at room temperature for 24 h to ensure optimum sensitizer uptake.

Counter electrode was prepared by drop casting process of platinum paste, onto FTO substrates ($2 \text{ cm} \times 1.5 \text{ cm}$, 8 ohm/square-Dyesol). After drying, this paste was gradually sintered at 450°C for 30 min, this results in uniformly distributed platinum nanoclusters, providing good transparency and high catalytic activity for the electrochemical reduction of I_3^- to I^- . A hole on the counter electrode was drilled for electrolyte injection.

For DSSCs assembling, low temperature thermoplastic sealant (purchased from Dyesol) as casket was utilized to sandwich a Pt-coated counter electrode and a dye-coated TiO_2 photoelectrode.

Finally, the electrolyte was injected into DSSC device through a hole, which was sealed by a piece of thermoplastic and a cover-class. The electrolytes solutions were consist of iodine (I^-) and triiodide (I_3^-) as a redox couple. The typical photoactive area of the finished cell was 0.19 cm^2 .

The J-V performances activities of the fabricated DSSCs were measured under solar simulator illumination (AM1.5 condition, 100 mW/cm^2). 450 W Xenon (Oriol) was utilized as source of light. J-V data collection using Keithley 2400 and LabView data conquest software photon-to-current conversion measurement IPCE was conducted with Enlitec.

Five different ionic liquids compound (3–7) samples were applied in DSSCs as Redox Mediator in this work. Electrolyte

solutions was prepared the combinations of 0.6 M ionic liquid + 0.1 M LiI + 0.05 M I_2 in MPN. The commercial iodide based redox electrolyte was the sixth sample used for comparison.

2.4. Synthesis of ionic liquids

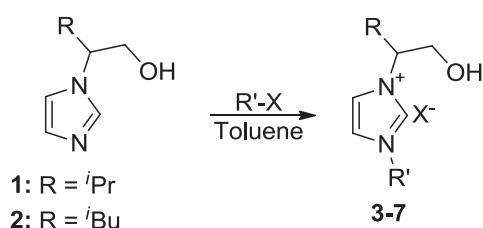
1-Substituted imidazoles (**1**, **2**) were obtained by the cyclocondensation of glyoxal, ammonium acetate, formaldehyde and commercially available amino alcohol, according to the previously published procedure [14]. The 1-substituted imidazoles (**1**, **2**) were then readily converted into the desired imidazolium salts (**3**–**7**) as shown in Fig. 1, which were obtained as viscous oils in high yields (80–94%). The intermediate compounds and the imidazolium salts were characterized by spectroscopic methods. The NMR data of 1-substituted imidazoles (**1**, **2**) and imidazolium salt (**4**) are consistent with the literature [14–16]. The ^1H and ^{13}C NMR spectra of the salts showed characteristic NCHN resonances between 9.57–9.78 ppm and 135.4–135.9 ppm, respectively. The signals due to the imidazole ring resonances were observed at 7.28–7.51 ppm. ^1H and ^{13}C NMR spectra of compounds were presented in Fig. 2.

3: ^1H NMR (400 MHz, CDCl_3 , TMS, 25°C , ppm): 0.83; 1.07 (6 H, d, $J = 8.0$ Hz, $\text{CH}(\text{CH}_3)_2$), 0.97 (3 H, t, $J = 8.0$ Hz, CH_3), 1.34–1.43 (2 H, m, CH_2), 1.87–1.95 (2 H, m, CH_2), 2.18–2.24 (1 H, m, $\text{CH}(\text{CH}_3)_2$), 3.93–4.06; 4.29–4.37 (5 H, m, CH_2OH , NCH_2 , NCH), 7.31; 7.40 (2 H, s, im-CH), 9.76 (1 H, s, NCHN). ^{13}C NMR (100 MHz, CDCl_3 , TMS, 25°C , ppm): 13.4, 19.1, 19.2, 19.3, 29.5, 31.9, 49.6, 61.0, 68.9, 121.9, 135.8. Anal. Calc. for $\text{C}_{12}\text{H}_{23}\text{BrN}_2\text{O}$: C, 49.49; H, 7.96; N, 9.62. Found: C, 49.48; H, 7.95; N, 9.60. Yield: 91%.

4: ^1H NMR (400 MHz, CDCl_3 , TMS, 25°C , ppm): 0.78; 1.03 (6 H, d, $J = 6.8$ Hz, $\text{CH}(\text{CH}_3)_2$), 0.83 (3 H, t, $J = 6.8$ Hz, CH_3), 1.21–1.29 (10 H, m, CH_2), 1.86–1.89 (2 H, m, CH_2), 2.18–2.24 (1 H, m, $\text{CH}(\text{CH}_3)_2$), 3.94–3.98; 4.26–4.34 (5 H, m, CH_2OH , NCH_2 , NCH), 7.39; 7.51 (2 H, s, im-CH), 9.68 (1 H, s, NCHN). ^{13}C NMR (100 MHz, CDCl_3 , TMS, 25°C , ppm): 13.9, 19.1, 19.3, 22.4, 26.0, 28.8, 28.9, 29.6, 30.0, 31.5, 49.9, 60.9, 69.1, 121.7, 122.1, 135.8. Anal. Calc. for $\text{C}_{16}\text{H}_{31}\text{BrN}_2\text{O}$: C, 55.33; H, 9.00; N, 8.07. Found: C, 55.32; H, 9.00; N, 8.05. Yield: 88%.

5: ^1H NMR (400 MHz, CDCl_3 , TMS, 25°C , ppm): 0.83; 1.05 (6 H, d, $J = 6.8$ Hz, $\text{CH}(\text{CH}_3)_2$), 0.87 (3 H, t, $J = 6.8$ Hz, CH_3), 1.24–1.33 (14 H, m, CH_2), 1.91–1.93 (2 H, m, CH_2), 2.18–2.24 (1 H, m, $\text{CH}(\text{CH}_3)_2$), 3.94–4.06; 4.27–4.36 (5 H, m, CH_2OH , NCH_2 , NCH), 7.28; 7.38 (2 H, s, im-CH), 9.78 (1 H, s, NCHN). ^{13}C NMR (100 MHz, CDCl_3 , TMS, 25°C , ppm): 13.9, 19.2, 19.3, 22.5, 26.1, 28.9, 29.1, 29.3, 29.6, 30.0, 31.7, 49.9, 60.9, 69.1, 121.7, 122.1, 135.9. Anal. Calc. for $\text{C}_{18}\text{H}_{35}\text{BrN}_2\text{O}$: C, 57.59; H, 9.40; N, 7.46. Found: C, 57.58; H, 9.39; N, 7.45. Yield: 80%.

6: ^1H NMR (400 MHz, CDCl_3 , TMS, 25°C , ppm): 0.84; 1.09 (6 H, d, $J = 8.0$ Hz, $\text{CH}(\text{CH}_3)_2$), 0.98 (3 H, t, $J = 8.0$ Hz, CH_3), 1.35–1.42 (2 H, m, CH_2), 1.89–1.97 (2 H, m, CH_2), 2.23–2.35 (1 H, m, $\text{CH}(\text{CH}_3)_2$), 4.01–4.05; 4.28–4.35 (5 H, m, CH_2OH , NCH_2 , NCH), 7.33; 7.43 (2 H, s, im-CH), 9.61 (1 H, s, NCHN). ^{13}C NMR (100 MHz, CDCl_3 , TMS, 25°C , ppm): 13.5, 19.3, 19.4, 29.6, 31.9, 49.9, 60.9, 69.1, 122.0, 122.2, 135.4. Anal. Calc. for $\text{C}_{12}\text{H}_{23}\text{IN}_2\text{O}$: C, 42.61; H, 6.85; N, 8.28. Found: C, 42.57; H, 6.83; N, 8.25 Yield: 97%.



Compound	R	R'	X
3	$i\text{Pr}$	C_4H_9	Br^-
4	$i\text{Pr}$	C_8H_{17}	Br^-
5	$i\text{Pr}$	$\text{C}_{10}\text{H}_{21}$	Br^-
6	$i\text{Pr}$	C_4H_9	I^-
7	$i\text{Bu}$	C_4H_9	I^-

Fig. 1. Synthesis of imidazolium salts (3–7).

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