



Influence of polyoxyethylene phytosterol addition in ionic liquid-based electrolyte on photovoltaic performance of dye-sensitized solar cells



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ABSTRACT

In this work, we studied influence of polyoxyethylene phytosterol (BPS) addition in ionic liquid (IL)-based electrolyte on photovoltaic performance of dye-sensitized solar cells (DSSCs) using 1-methyl-3-propylimidazolium iodide as an IL. Surface tension, photocurrent density-voltage characteristics and electrochemical impedance spectra were measured to clarify the role of BPS in the DSSCs using three different dyes. The results showed that the IL solution of less solvophilic BPS-EO5 exhibited a better surface active property and a weaker dye-desorption effect than BPS-EO30 and BPS-PO7/EO30. Short-circuit current densities of the N719- and NKX2677-sensitized cells were found to be noticeably increased by the addition of either BPS-EO5 or BPS-EO30 to the IL-based electrolyte in the concentration range of 0.001–0.01 mol dm⁻³. Enhanced photovoltaic conversion efficiencies were obtained for these DSSCs, which most likely resulted from the effects of BPS on reducing charge-transfer resistance at the TiO₂/dye/electrolyte interface and on increasing electron lifetime within the TiO₂ photoanode.

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1. Introduction

Dye-sensitized solar cells (DSSCs) have been attracting attention as candidates for next generation of solar cells because of their advantages of low production cost, easy methods of fabrication and relatively high-energy conversion efficiency [1,2]. The conventional DSSC consists of a dye-sensitized nanoporous TiO₂ photoanode, a platinum cathode and a liquid electrolyte. The liquid electrolyte encapsulated in the gap between the electrodes is usually composed of organic solvents, I⁻/I₃⁻ redox couple and some additives. However, use of volatile organic solvents such as nitrile derivatives causes vaporization and leakage of the liquid electrolyte especially at elevated temperatures, resulting in a poor durability for the long-term operations.

Among options being investigated to avoid this problem, electrolytes based on room-temperature ionic liquids (ILs) are some of the most interesting choice for their advantages not only negligible vapor pressure at ambient temperatures, but also high ionic conductivity, non-flammability and outstanding electrochemical stability [3]. These unique properties of ILs have been considered to be beneficial for the practical applications of DSSCs [4,5]. However, ILs of imidazolium compounds, which are most

widely used as an electrolyte solvent for the DSSCs, generally have a serious drawback of intrinsic high viscosity, being responsible for significantly small diffusion coefficients of the redox couple in the IL-based electrolytes [6–8]. Compared to the cells based on conventional organic solvent-based electrolytes, higher recombination and lower injection of charge promote back electron transfer, leading to lower efficiencies of the IL-based cells [9]. Even though Grotthuss-like fast charge transport based on exchange reaction of the I⁻/I₃⁻ redox couple occurs in the IL [6,10], the photovoltaic performances of these DSSCs still remain insufficient. Similarly to the organic solvent-based electrolytes, addition of 4-*t*-butylpyridine (TBP) in the ILs improves cell performance to some extent [11–13], nevertheless volatility of TBP makes it unfavorable for long-term usage [14].

Phytosterols involving β-sitosterol, campesterol, stigmasterol and tocopherol are plant-based sterol derivatives, being attracted much attention because of their potential health benefits. Ethoxylation of the phytosterols generates surfactants suitable for cosmetics with low toxicity and biocompatible properties [15]. It has been reported that the phytosterol surfactant (BPS) shows excellent interfacial properties and self-assembling behavior even in ILs [16–18]. Consequently, BPS is expected to have a potential for DSSC applications as an additive in the IL-based electrolyte capable of modifying the surface of working electrode through adsorption on the TiO₂ particles. Depending on the BPS concentration, the adsorption layer may change the interfacial properties between

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TiO₂ and IL, and thus influencing the photovoltaic characteristics of the cell.

Although many works have been reported on the DSSC using IL-based electrolytes so far, there are few researches on the effects of addition of surfactant such as BPSs to the IL-based electrolyte. Therefore, on the basis of the motivation stated above, we used three types of BPSs with different solvophilic moiety (BPS-EO5, BPS-EO30 and BPS-PO7/EO30) and explored their effects on the photovoltaic performance of the DSSC using IL-based electrolyte. Surface active properties of BPSs were evaluated from surface tension measurements of the BPS-containing IL solutions, and characteristics and performance of the DSSCs using various IL-based electrolytes were examined via photocurrent density (*I*)-voltage (*V*) curves and electrochemical impedance spectroscopy (EIS). The measurement of photovoltaic characteristics was carried out for each cell sensitized with three different dyes, and the results obtained for these cells were compared and discussed.

2. Experimental details

2.1. Materials

1-methyl-3-propylimidazolium iodide (MPII) was purchased from Tokyo Chemical Industry and used as an IL solvent for the electrolyte throughout the study. BPSs with different polyoxyethylene or polyoxypropylene-polyoxyethylene chain (BPS-EO5, BPS-EO30 and BPS-PO7/EO30) were supplied by Nikko chemicals and used as surfactants without further purification. Chemical formulae of these materials are presented in Fig. 1. Three types of commercially available dyes, di-tetrabutylammonium cis-bis(isothiocyanato)bis(2,2'-bipyridyl-4,4'-dicarboxylato)ruthenium(II) (N719, Peccell Technologies), 2-cyano-3-[5'-(1,1,6,6-tetramethyl-10-oxo-2,3,5,6-tetrahydro-1H,4H,10H-11-oxa-3a-aza-benzo[de]anthracen-9-yl)-[2,2']bithiophenyl-5-yl]acrylic acid (NKX2677, Hayashibara Biochemical Laboratories) and 2-cyano-5-[4-(dimethylamino)phenyl]-2,4-pentadienoic acid (NKX2553, Hayashibara Biochemical Laboratories), were adopted as sensitizers in DSSCs (Fig. 2) and used as received. TiO₂ powder (P25) was obtained from Nippon Aerosil Co., Ltd. All other chemicals are commercially available.

2.2. Methods

Nanoporous TiO₂ electrodes were prepared on conductive fluorine-doped SnO₂ (FTO) substrates (A110U80, Asahi Glass) by electrophoretic deposition technique. The TiO₂ particles were deposited from a 2.2 wt% TiO₂ suspension in 50 vol% aqueous ethanol applying a DC voltage of 10V for 35 sec with an inter electrode distance of 5 mm. Under these conditions, density of the deposited TiO₂ could be adjusted to 15 g m⁻² (11 μm thick) at which the highest cell performance was obtained in our previous

study [19]. The resulting electrodes were sintered at 500 °C for 30 min in air and afterwards the TiO₂ film was scraped off so that only an ϕ 8 mm circle of the film was left on the FTO substrate. For sensitization, the electrodes were heat-treated again at 500 °C for 10 min, followed by immersing in a *t*-butanol/ethanol/acetonitrile (1:1:2) solution of 3.0 × 10⁻⁴ mol dm⁻³ sensitizing dye at 40 °C for 2 h. Then, the electrodes were rinsed with acetonitrile and dried in air. Conductive FTO substrate coated with sputtered Pt film was used as a counter electrode. The dye-sensitized TiO₂ electrode and the counter electrode were assembled in a sandwich-type cell by sealing together with a 50 μm thick hot-melt gasket (Surlyn, DuPont). The gap between the electrodes was filled with the IL-based electrolytes consisting of 0.3 mol dm⁻³ I₂, 0.5 mol dm⁻³ LiI and 0–0.1 mol dm⁻³ BPS in MPII by injecting the electrolyte before the sealing. A schematic illustration of the DSSC structure is shown in Fig. S1 in the online version at DOI: <http://dx.doi.org/10.1016/j.electacta.2016.10.045>.

I-*V* curves were measured with a digital source meter (Keithley model 2400, Keithley Instruments) using a solar simulator (PECL10, Peccell Technologies) with simulated sunlight of AM 1.5 (100 mW cm⁻²). Data were recorded after 4-cycle scans applying a black mask with a circular window (active area: 24 mm²) on the TiO₂ film side. To suppress hysteresis between forward and backward scan, the scan rate was set at a relatively slow value of 0.001 V s⁻¹. *I*-*V* characteristics were also observed in the dark at the same scan rate. EIS spectra of the DSSCs were obtained by a potentiostat (VersaSTAT 3-100, Princeton Applied Research) under illuminating condition. The spectra were scanned in the frequency range of 10⁻²–10⁶ Hz at room temperature with applied bias voltage and AC amplitude set at open circuit voltage (*V*_{oc}) and 10 mV, respectively.

Desorption characteristics of the sensitizing dyes in the presence of BPS in IL were examined by immersing the dye-adsorbed TiO₂ film in 2 ml MPII containing various concentrations of BPS for 24 h. The TiO₂ film was subsequently taken out from the IL solution, rinsed with acetonitrile and dried. Amount of desorbed dye from the TiO₂ film was difficult to estimate from absorption spectrum of the resultant MPII solution because the trace amounts of impurities often cause discoloration of IL [20], which are usually yellow-colored. Therefore, to obtain an indication of the amount of dye remaining in the TiO₂ film, we instead measured ultraviolet-visible (UV-vis) diffuse reflectance spectra of the dye-adsorbed TiO₂ film using a Shimadzu UV-3100PC spectrophotometer equipped with an integrating sphere attachment (ISR-3100, Shimadzu).

According to the Wilhelmy plate method, equilibrium surface tension measurements were carried out for the MPII solutions containing various concentrations of BPSs by a surface pressure meter (HBM-AP type, Kyowa Kaimenkagaku) at 20 °C. Owing to high viscosity of the solutions, sufficient waiting time (1–48 h) was required to ensure adsorption equilibrium for each measurement.

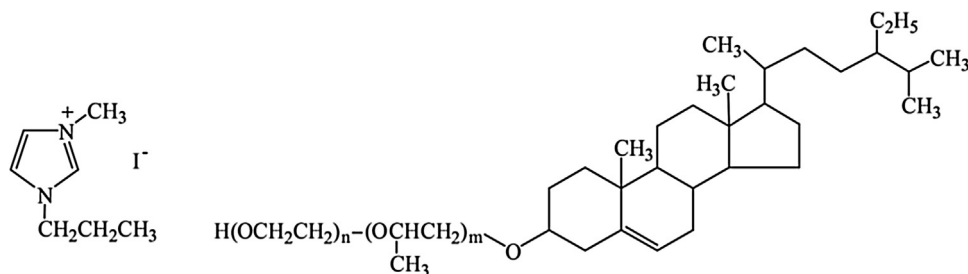


Fig. 1. Structural formulae of MPII (left) and BPS-PO_m/EO_n (right); BPS-EO5 (*m*=0, *n*=5), BPS-EO30 (*m*=0, *n*=30), BPS-PO7/EO30 (*m*=7, *n*=30).

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