



A dual function of high performance counter-electrode for stable quasi-solid-state dye-sensitized solar cells



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HIGHLIGHTS

- PEDOT–PAA–PSS is prepared and used as counter electrode in dye-sensitized solar cell.
- The hybrid can store liquid electrolyte and has good electrocatalytic activity.
- The efficiency of the solar cell using the dual function electrode reaches 6.35%.

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ABSTRACT

A dual function polymer hybrid material poly(3,4-ethylenedioxythiophene) (PEDOT)–poly(acrylic acid) (PAA)–poly(styrenesulfonate) (PSS) was fabricated both as a counter electrode (CE) for I_3^- reduction and for storage of the liquid electrolyte in quasi-solid-state dye-sensitized solar cells (QS-DSSCs). To enhance the electrocatalytic activity of the nano-PEDOT CE and to improve the ionic conductivity of the three-dimensional (3D) PAA–PSS network, the columnar PEDOT was electropolymerized through the 3D PAA–PSS network. The QS-DSSC showed a high photovoltaic conversion efficiency of 6.35% under full sunlight illumination (100 mW cm^{-2} , AM1.5 G), which reduced to 6.03% after 100 days, indicating this QS-DSSC had a relatively good long-term stability.

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1. Introduction

Since the first publication on dye-sensitized solar cells (DSSCs) by O' Regan and Grätzel in 1991 [1], the cells have attracted a great deal of attention due to their low cost, simple preparation procedures, and relatively high energy conversion efficiency [2–4]. A typical DSSC consists of a dye-sensitized TiO_2 photoanode, a redox electrolyte, and a platinumized counter electrode (CE). The CE should catalyze the reduction of I_3^- to I^- . To date, poly(3,4-ethylenedioxythiophene) (PEDOT) [5–8] has been investigated as a stable, highly conductive and more cost-effective CE replacing platinum [9] since Yohannes and Inganäs [10] reported that doped PEDOT can catalyze the I_3^-/I^- redox couple.

The DSSC electrolyte provides internal ionic conductivity within the porous nanocrystalline TiO_2 film [11] and is an important factor in determining the cell performance. Despite the high conversion efficiencies achieved for the DSSCs with liquid electrolytes, potential problems such as the leakage and volatilization of organic solvents in liquid electrolytes, are considered as some of the critical factors limiting the long-term performance and practical use of DSSCs [12–14]. Thus, all-solid-state electrolytes [15–18], ionic liquid electrolytes [19,20], and quasi-solid-state electrolytes [12,13,21–24] have been proposed as alternatives to liquid electrolytes. Among them, polymer gel electrolytes are considered as a substitute for liquid electrolytes, due to their high ionic conductivity, good interfacial filling properties, and relatively high long-term stability [25]. Cells using these electrolytes may be termed quasi-solid-state dye-sensitized solar cells (QS-DSSCs). Recently, polymer gel electrolytes based on a super-absorbent polymer poly(acrylic acid) (PAA) have been reported [13]. In it, either the carboxyl groups are modified or amphiphilic groups (e.g.,

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polyethylene glycol [12,13], glycerin [26], *g*-cetyltrimethyl ammonium bromide [27], or gelatin [28]) are introduced to improve organic solvent absorbency. Introduction of conducting polymers (e.g., polypyrrole [22] or polyaniline [24]) enhances electron conductivity in the polymer hybrid. Such polymer hybrids showed excellent stability and liquid electrolyte absorbent ability, and were used to form high performance, stable QS-DSSCs.

In this paper, we report a novel dual function organic polymer hybrid material PEDOT–PAA–poly(styrenesulfonate) (PSS) prepared using pulse potentiostatic electropolymerization of high-uniformity nano-PEDOT film on a fluorinated tin oxide (FTO) substrate. It was then subjected to sequential chemical redox polymerization of a microporous PAA–PSS thin film, which was used as the CE for I_3^- reduction and for storage for the liquid electrolyte in the QS-DSSC. The latter achieved the relatively high energy conversion efficiency of 6.35%, which fell to 6.03% after 100 days, showing that this QS-DSSC had a relatively good long-term stability.

2. Experimental

2.1. Fabrication of CEs

Pulse potentiostatic electropolymerization [7,8] of PEDOT onto cleaned FTO glass substrates ($13 \Omega \square^{-1}$, NSG) were carried out using a Zahner electrochemical workstation (Germany) from a simple three compartment deposition bath containing 2.0 mM 3,4-ethylenedioxythiophene monomer (99% EDOT, ARCOS), 10 mM poly(styrenesulfonate) (PSS), and 10 mM lithium perchlorate (LiClO_4) in aqueous solution (called here EDOT aqueous solution) at ambient atmosphere. Preparative parameters for PEDOT CEs were selected as 1.2 V imposed pulse potential, 0.2 V pulse-reversal potential, 1 s pulse period, 0.5 s pulse-reversal period, and total 500 s duration time. Cleaned FTO glass substrates and a Pt wire were used as working electrodes and as the counter electrode, respectively, with a saturated silver/silver chloride (Ag/AgCl) reference electrode. The PEDOT CEs thus obtained were rinsed in distilled water and dried under a cool air flow.

The oligo–PAA–PSS hybrid was prepared by the following processes. First, 10 ml acrylic acid monomer, 3.0 g PSS, with 0.01 g *N,N'*-methylenebisacrylamide as cross-linker agent were dissolved in 12 ml deionized water with stirring. Second, 0.1 g ammonium persulfate used as a redox initiator for the PAA polymerization reaction was added drop-wise into the above mixture at 4 °C under vigorous stirring to form homogeneous oligo–PAA–PSS hybrid polymer.

The product was coated onto the PEDOT CEs to form PEDOT–PAA–PSS (further abbreviated as P-0) CEs by the doctor blade technique. Polymerization was allowed to take place at 80 °C for 30 min under nitrogen atmosphere. The resultant was washed at

room temperature in excess deionized water to remove impurities, and vacuum-dried at 80 °C for 2 h. P-0 CEs were immersed in the above EDOT aqueous solution for 6 h. Electropolymerization of PEDOT onto the P-0 CE was then carried out using the method described for FTO over periods of 500, 1000, and 1500 s. The P-0, P-500, P-1000, and P-1500 CEs obtained were rinsed in deionized water to remove any impurities and freeze-dried (FD-1A-50, China).

2.2. Characterization

The surface features of samples were observed using scanning electron microscopes (SEM, JSM-7600F and S-3500N). Fourier transform infrared spectra (FTIR) of samples were recorded on a Nicolet Impact 410 FTIR spectrophotometer (UK) with KBr cut-off. Cyclic voltammogram (CV) for the I^-/I_3^- system used an electrolyte consisting of 50 mM LiI, 10 mM I_2 , and 500 mM LiClO_4 dissolved in a 2:8 by volume mixture of *N*-methyl-2-pyrrolidone (NMP, ARCOS) and 3-methoxypropionitrile (3-MPN, ARCOS). Results were obtained in the potential range -0.4 V to 0.4 V at a scan rate of 10 mV s^{-1} using a computer-controlled potentiostat (PGSTAT320N, Autolab). A three-electrode electrochemical cell was used, containing the CEs obtained as working electrodes, a Pt-foil counter electrode, and a Pt wire reference electrode. The symmetrical dummy cells (shown in Fig. 1a–c) were used for the electrochemical impedance spectroscopy (EIS) and Tafel polarization measurements. The EIS measurements were conducted by using the Zahner electrochemical workstation. The impedance studies were carried out simulating open-circuit conditions in the dark under ambient atmosphere, and the impedance data covered a frequency range of 10^{-1} – 10^6 Hz with zero bias potential, and at 10 mV signal amplitude. The resulting impedance spectra were analyzed by means of Z-view software. Tafel polarization curves were measured using an electrochemical workstation system (CHI660D, China) at a scan rate of 10 mV s^{-1} .

The redox electrolyte used for EIS, Tafel polarization, and photovoltaic conversion measurements consisted of 1 M 1,3-dimethylimidazolium iodide (Merck), 0.5 M 4-tert-butylpyridine (Aldrich), 0.15 M iodine (J.T. Baker), and 0.1 M guanidine thiocyanate (Aldrich) in a 2:8 NMP and 3-NMP mixture. The CV, EIS, Tafel polarization, and photovoltaic conversion measurements were obtained after the samples were immersed in the corresponding test solution for 24 h to absorb electrolytes adequately.

2.3. Measurement

The absorbency (g g^{-1}) of each sample was measured according to the equation below [29]:

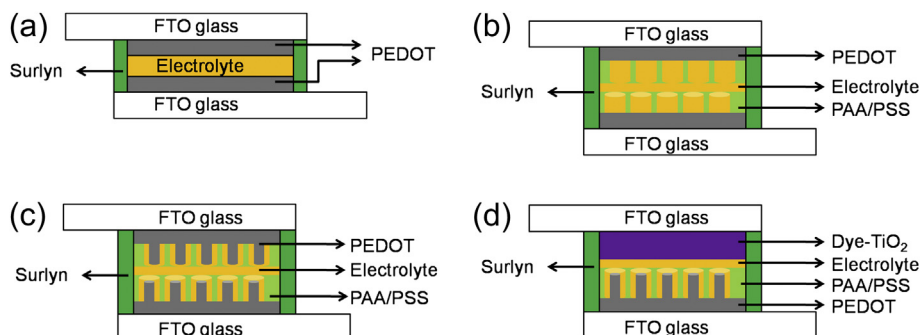


Fig. 1. Symmetric dummy cells (a, b, and c) used for EIS and Tafel polarization measurements, schematic diagram of the QS-DSSC (d).

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