



Influence of electropolymerized polypyrrole optical properties on bifacial Dye-Sensitized Solar Cells

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ABSTRACT

In this work the effect of different doping anions on transparency, photovoltaic efficiency and interface properties of electropolymerized polypyrrole (PPy) films as counter electrode in bifacial DSSCs was study. The transparency of the counter electrode becomes relevant in order to enhance the final device efficiency and for alternative application area like of Building-Integrated Photovoltaic. PPy was prepared by electrochemical process with doping anions: chloride, perchlorate, sulfate and dodecylbenzenesulfonate. PPy doped with high concentration (0.5 M) of small anions (chloride) produced the highest transparency film (65% at 525 nm), the DSSCs with highest efficiency ratio (close to 70%) and lowest device reflectance (11%). Considering all of these results, not only the transparency of counter electrode was found to influence the efficiency of the bifacial DSSCs, but also the optical properties of PPy/electrolyte interface. This interface is probably strictly affected to the PPy morphology produced by different doping anions and the electropolymerization process used.

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1. Introduction

Recently, much attention has been paid on Dye-Sensitized Solar Cells (DSSCs) as a promising photovoltaic technology formed by eco-friendly components [1–6]. Building-Integrated Photovoltaic (BIPV) in particular, can boost the transition to market for the DSSCs. One of the more desirable characteristics of DSSCs for BIPV in fact is to be bifacial, and therefore convert light coming from both side of the cell, i.e. from outdoor and indoor. For this purpose however, both the photoanode and the counter electrode must be highly transparent [7,8]. This property is also required in order to increase the incident light harvesting and therefore enhance the overall conversion efficiency [9–11]. In fact, if the sunlight irradiation comes simultaneously from the front and the rear side of the cell more dye molecules are excited enhancing the short-circuit current density and the efficiency of the entire cell. Finally, bifacial DSSCs thanks to their advantage of higher light-harvesting and their capabilities of utilizing the incident light from both side, could help to further bring down the cost of energy production [12]. Traditionally, Platinum is used as transparent DSSCs counter

electrode but alternative materials must be nowadays considered. Platinum is expensive, scarce in nature and shows stability problem with the iodide/triiodide electrolyte in organic solvent used in the most common DSSC architecture. Moreover is not so effective when other redox couples such cobalt-complex, T_2/T^- (transparent electrolyte) are considered [13–15]. Literature reports a lot of possible alternative materials, such as carbon materials (carbon black, nanotube, graphene), polymer-carbon composite, transition metal compounds and conducting polymers (polyaniline, polythiophene, polypyrrole) [16–19]. As described before, efficient bifacial DSSCs requires an highest transparent counter electrode. Zhao et al. [20] achieved a conversion efficiency of 5.04% and 6.07% in rear and front side illumination condition respectively using a transparent carbon counter electrode prepared by carbonization. On the other hand, metal selenide alloys of Cobalt, Nickel and Iron were able to increase the final device efficiency of carbon based one [21–23]. Also conducting polymer like poly (3,4-ethylenedioxythiophene) (PEDOT) and polyaniline (PANI) and hybrid compounds with graphene were successfully used to obtained an high efficient bifacial DSSCs [9,10,24,25]. PEDOT film in particular was able to produce bifacial DSSCs with efficiency of 6.35% and 4.98% (front and rear side illumination) while comparable results were obtained for PANI counter electrodes (6.54% and 4.26%) [9,24]. Recently, polypyrrole (PPy) has received a lot of attention as a promising alternative to

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Platinum counter electrode for traditional and bifacial DSSCs due to its high conductivity, facile synthesis, low cost, high catalytic activity for I_3^- reduction [26–32]. PPy can be easily prepared by chemical oxidation or electrochemical oxidation of pyrrole monomer. The second one is usually preferred due to controllable initiation and termination of polymerization, easy and in situ polymerization at low temperature, controllable film thickness (controlling the amount of charge that flow inside the electrode) and amount of doping anions that is possible to include in the polymeric chain [27,33,34]. Zhang et al. [27] reports a study about the influence of doping anions on the properties of electropolymerized PPy as counter electrode in traditional DSSCs where the film doped with dodecylbenzenesulfonate anions shows the photovoltaic conversion efficiency up to 5.40%. To best of author knowledge, there are no reports regarding the influence of other doping anions on the properties of electropolymerized PPy used as counter electrode in bifacial DSSCs and how these anions can influence the final efficiency. So, the aim of this work was to investigate the influence of doping anions concentration and chemical properties on transmittance, catalytic activity and photovoltaic efficiency of PPy based DSSCs. In particular, these properties were evaluated in bifacial devices in order to understand which parameters control the working mechanisms of these cells and therefore their efficiencies. Four different doping anions were considered: potassium chloride, lithium perchlorate, sulphuric acid and sodium dodecylbenzenesulfonate.

2. Experimental

2.1. Preparation of electropolymerized PPy and Platinum electrodes

Electropolymerization of pyrrole (98% Sigma Aldrich) was performed in potentiostatic condition of 1 V vs SCE for different times with an Autolab PGSTAT302N (Metrohm, the Nederland). The Fluorine doped Tin Oxide coated glasses (FTO, sheet resistance 7 Ω /sq. Sigma-Aldrich) were pre-cleaned before deposition using isopropyl alcohol in an ultrasonic bath for 15 min. The electropolymerization was carried out in a conventional three-electrodes electrochemical cell with FTO (6.25 cm² substrate with deposition area of 0.25 cm²) as working electrode, a Platinum plate as auxiliary electrode and SCE (AMEL electrochemistry, Italy) as a reference. The polymerization was performed under stirring condition in an aqueous solution (Milli-Q water, >18 M Ω cm) containing 0.05 M pyrrole with different concentrations of doping salts of potassium chloride (Merck), lithium perchlorate (Sigma Aldrich), sulphuric acid (96% Sigma Aldrich) and sodium dodecylbenzenesulfonate (Sigma Aldrich) respectively. Doping anions concentration were changed between 0.2 M (low concentration) and 0.5 M (high concentration). After polymerization, the obtained PPy films named (Cl^- , ClO_4^- , SO_4^{2-} and DBS^-) were rinsed with Milli-Q water in order to remove the unreacted pyrrole monomer and dried in air for one night. As a reference, a Platinum electrode were also prepared by sputtering process (Q150T S/E/ES Quorum Technologies Ltd., U.K.) and thermal treatment at 450 °C for 30 min.

2.2. Depositions of TiO_2 photoanodes and DSSCs assembly

TiO_2 screen printing inks were deposited onto FTO glass substrates using a semi-automatic screen-printer (AUR'EL 900, AUR'EL Automation s.p.a., Italy), and treated at 450 °C for 30 min. The thickness of the films was adjusted to reach 8 μ m. In the samples used for the final devices the substrates were coated with a TiO_2 blocking layer (BL), according to the procedure reported by Sangiorgi et al. [35]. After sintering, the films were immersed in a 50 mm $TiCl_4$ aqueous solution at 70 °C, and then fired at 450 °C for

30 min. The as-obtained photoanodes were then dipped for 16 h in a 0.3 mM ethanolic solution of (*cis*-diisothiocyanato-bis (2,2'-bipyridyl-4,4'-dicarboxylato) ruthenium (II) bis (tetrabutylammonium) (N719 dye, Sigma-Aldrich). A pre-drilled FTO coated glass covered with a sputtered Pt layer or electropolymerized PPy film was used as counter electrode. After the photoanode sensitization the electrodes were assembled into a sandwich type cell and sealed with a hot melt gasket of Meltonix (thickness 25 μ m, Solaronix, Switzerland). Electrolyte (Iodolyte Z100, Solaronix, Switzerland), was introduced in the cell via vacuum back filling through the hole in the counter electrode. Finally the hole was sealed using a Meltonix film and a small cover glass. The active area of the solar cells was fixed at 0.25 cm².

2.3. Films and DSSCs characterizations

Transmittance spectra of the PPy films in the UV-Vis range were recorded on PVE300 system (Bentham, United Kingdom). The morphologies of the PPy films were observed by field emission gun-scanning electron microscope (FE – SEM, SIGMA, Zeiss, Germany). The catalytic activity of PPy film for the $3I^-/I_3^-$ redox reaction was evaluated by cyclic voltammetry (CV) in an acetonitrile solution containing 0.010 M I^- (Sigma Aldrich), 0.001 M I_2 (Sigma Aldrich) and 0.1 M $LiClO_4$ (Sigma Aldrich). The working electrode was an FTO/PPy or FTO/Pt, the reference was SCE and auxiliary a Platinum plate. The CV analysis were performed in the potential range between ± 1 V vs SCE at different scan rate and prior to each analyses the solutions were purged with N_2 for 10 min. Moreover, the Tafel polarization tests on $3I^-/I_3^-$ redox reaction were done in the potential range between ± 1 at scan rate of 50 mV/s. For this analysis, a symmetric cells with two identical PPy or Platinum electrodes with an active area of 0.25 cm² were assembled with the same electrolyte used in the DSSCs. In order to evaluate the DSSCs performance, J-V curves in front- and rear-illumination conditions were done under AM 1.5 simulated illumination with power density of 1000 W m⁻² (calibrated with a standard silicon cell). The J-V curves were measured using an Abet Technologies solar simulator (SUN 2000, USA) and a Keithley 2400 source/meter (Keithley, USA). In order to understand the influence of the PPy optical properties on the efficiency ratio of bifacial DSSCs, the reflectance spectra of complete cells were acquired by PVE300 system with integrating sphere. Electrochemical Impedance Spectroscopy (EIS) of Pt and PPy based DSSCs was also performed in the frequency range from 10⁵ to 0.05 Hz, with amplitude of 10 mV, in the dark and open circuit conditions. For the fitting procedure Z-View (Scribner Associates Inc.) software was used. All of these electrochemical characterizations were done by Autolab PGSTAT302N and Nova 1.10 (Eco Chemie, the Nederland) was used as software to analyse the obtained data. The graphical elaborations of the experimental values were made using OriginLab Pro 2015 (USA).

3. Results and discussion

3.1. Influence of doping anions concentration on morphology and transmittance

In order to study the influence of the anions on the PPy transparency, it is essential to compare films with the same thickness. During the electropolymerization process, the charge that flow through the FTO electrode was determined and used to determine the amount of PPy deposited on the substrate. The mean film thickness was therefore estimated from the electrical charge associated to the pyrrole oxidation considering the Faraday's law, and assuming 100% current efficiency of PPy formation [29]. So, the PPy films doped with different anions were

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