APPLICATIONS OF PProDOT-Bz₂/PT COMPOSITE COUNTER ELECTRODES IN DSSCS UNDER DIFFERENT ILLUMINATION INTENSITIES

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ABSTRACT

Poly(3,3-dibenzyl-3,4-dihydro-2H-thieno[3,4-b][1,4]dioxepine) (PProDOT-Bz₂)/platinum (Pt) composites films were employed as counter electrodes (CE) for dye-sensitized solar cells (DSSCs). The composite films were deposited onto fluorine-doped tin oxide (FTO) substrate by sputtering Pt for 10 s and 30 s (Pt-FTO glass). 25 mC cm⁻² and 50 mC cm⁻² PProDOT-Bz₂ were electrochemically synthesized on 10 s and 30 s Pt-FTO glass. The composites films were characterized via electrochemical impedance spectroscopy (EIS). The DSSC based on 25 mC cm⁻² PProDor-Bz₂/30 s Pt composite films exhibits short-circuit current density (J_{sc}) of 11.11 mA cm⁻², open circuit voltage (V_{oc}) of 0.62 V, fill factor (FF) of 68.74%, and the photovoltaic conversion efficiency of 4.72%. Furthermore, the DSSC is a promising device because it can work under indoor fluorescent light. In this study, the composites counter electrodes under different light intensities are also investigated.

Keywords: Electrosynthesis, counter electrode, light intensity, PProDOT-Bz₂.

1. INTRODUCTION

Dye-sensitized solar cells (DSSCs) have become increasingly attractive due to their low cost and easy fabrication, after the report revealed by O'Regan and Grätzel in 1991 (O'Regan and Grätzel 1991). The CE of DSSCs is part of an important component in DSSCs. Meanwhile, Pt has superior electrochemical catalytic activity, good corrosion resistance, small resistance, large electrochemical active area and high reflecting properties, and so it has been regarded as suitable CE material. However, platinum-based CE is too expensive to be applied to DSSCs. With this in mind, research into this has been conducted to explore several possible substitutes. This is to substitute the CE material for platinum in DSSCs. Carbon-based materials, such as graphite, carbon black (Kay and Grätzel 1996; Liu *et al.* 2016), carbon nanotubes (CNTs)

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(Vijayakumar et al. 2017), carbon nanorods (CNRs) (Wang et al. 2017), hard carbon spheres (Bu and Kao 2017), graphene (Ma et al. 2017; Ghasemi et al. 2017; Zhou et al. 2016; Nemala et al. 2017; Kavan et al. 2016; Yuan et al. 2017; Miao et al. 2017) and carbon material fabricated from sucrose and glucose, etc., are considered to have great potential to replace Pt as counter electrode in recent years. They are low-cost and exist in large numbers. Also they have high catalytic activity and remain good chemical stability (Kang et al. 2016; Kumar et al. 2017). More importantly, conducting polymers are suitable to replace Pt as CEs due to their low-cost, excellent catalytic activity, and good conductivity. These properties are believed to have great potential to fabricate large-area CEs. In the past few years, conducting polymers, e.g. polyaniline (PANI) (Li et al. 2008; Li et al. 2009; Zhang et al. 2010), polypyrrole (PPy)(Bu et al. 2013; Veerender et al. 2012; Towannang et al. 2012), poly-3-methyl-thiophene (P3HT)(Torabi et al. 2014), poly (3,4-ethyl-enedioxythiophene)(PEDOT) (Belekoukia et al. 2016; Li et al. 2017; Kim et al. 2016), PEDOT:PSS (Li et al. 2013; Yue et al. 2015) and 3,4-propylenedioxythiophene derivatives (PProDOT derivatives) (Yeh et al. 2011) have been reported as promising CEs of DSSCs. Among them, PP or DOT derivatives have good catalytic properties and high specific surface areas. M.H. Yeh et al. have prepared PProDOT-Et₂/10 s Pt as CEs. The power conversion efficiency of DSSCs with the PProDOT-Et₂/10 s CE is 6.65%, which is higher than that of Pt CE with sputtering time for 720 s (6.43%) (Yeh et al. 2011). The improvement of photoelectric conversion efficiency for PProDOT- Et₂/Pt-based CE was ascribed to PProDOT-Et₂, for this showed a markedly high electrochemical surface area for the reduction from I3⁻ to I⁻(Yeh et al. 2011). In this study, we synthesized a novel polymer of PProDOT-Bz₂ electrochemically and prepared the PProDOT-Bz₂/ Pt composite film, by combining Pt with different sputtering time and PProDOT-Bz₂ with different charge capacity to improve the photoelectric conversion efficiency.

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2. EXPERIMENTAL

2.1 Materials

TiO₂ paste (ETERDSC Ti-2105) and (ETERDSC Ti-2325) were provided by Eternal Materials Co., Ltd., Taiwan. Ruthenium-535 (N3), iodine (I₂), lithium iodide (LiI), 4-tert-butylpyridine (TBP), absolute ethanol, and 1-propyl-2,3-dimethylimidazolium iodide were ordered from TCI company, UniRegion Bio-Tech, Katayama Chemical, Sigma-Aldrich, and Riedel-deHaen, and they were used as received. FTO substrates were obtained from C.P. Solar, Co., Ltd.

2.2 Synthetic procedure of ProDOT-Bz₂ monomer

1 g (0.007 mol) of 3,4-dimethoxythiophene, 1.37 g (0.0054 mol) of 2,2-dibenzy-1,3-propanediol, 0.04 g (0.0002 mol) of p-toluenesulfonic acid, and 50 mL of toluene, were combined in a round-bottomed flask. The solution was heated at 110°C under nitrogen for 2 days. The product was purified by flash column chromatograph along with silica gel used as stationary phase and dichloromethane/hexane (volume ratio of 1:2) used as the mobile phase. ProDOT-Bz₂ was a white powder and the yield was 60%. 1H NMR (700 MHz, CDCl₃, ppm): δ7.27 (m, 10H, phenyl-H), δ6.49 (s, 2H, Th-H), δ3.85 (s, 4H, OCH₂-), δ2.85 (s, 4H, phenyl-CH₂-).

2.3 Fabrication of PProDOT-Bz₂ counter electrodes

Figure 1 (a) provides schematic procedures of the preparation of PProDOT-Bz₂/Pt composite counter electrode. The FTO glasses were sequentially cleaned via sonication in acetone, ethanol, and D. I. water. The various Pt-FTO substrates were fabricated by sputtering Pt for 10s and 30 s onto FTO glass. The sputtering parameters of Ar flow rate, working pressure, and ration frequency sputtering power kept 10 sccm, 30 mTorr, and 60 W. The electrochemical system was mainly comprised of three electrodes, including various Pt-FTO glass substrates as working electrodes, Pt wires as counter electrodes, and Ag /AgCl as reference electrodes. The PProDOT-Bz₂ films were electro synthesized by potentiostatically at the potential of 1.4 V on Pt-FTO glass substrates using 0.1 M LiClO₄ and 6 mM ProDOT-Bz₂ in CH₃CN solution. The charge density was fixed at 25 or 50 mC cm⁻², respectively. Then, the prepared films were heated at 70°C for 1 h to remove the solvent.

2.4 Assembly of the DSSCs

The 12 μ m double-layer TiO₂ film was comprised of a transparent layer with a thickness of 8 μ m (ETERDSC Ti-2105), combined with a light scattering layer with a thickness of 4 μ m (ETERDSC Ti-2325) being coated in the cleaned FTO substrates by using the doctor blade method. Each layer was sintered in air at 500°C for 30 min. After sintering, the photoanode was immersed into the dye solution containing 0.3 mM N3 dye in ethanol for one day at room temperature. Finally, the TiO₂ photoanodes were coupled with various counter electrodes and electrolyte by a sandwiching process. The electrolyte solution was observed to contain 0.5 M lithium iodide, 0.05 M iodide, 0.5 M 4-tert-butylpyridine (TBP) and 0.6 M 1-propyl-2,3-dimethylimidazolium iodide (DMPII) in 3-methoxypropionitrile/acetonitrile (volume ratio of 1:1). The active area of the DSSCs was 0.25 cm². The components of the DSSCs are shown in Fig. 1 (b).

3. RESULTS AND DISCUSSION

3.1 Photovoltaic performances

The J-V curves of DSSCs based on different CEs under 100 mW cm⁻² light illumination are presented in Fig. 2, and the photovoltaic parameters are summarized in Table 1. As shown in Table 1, it could be described that DSSC based on PProDor-Bz₂/Pt composites CEs had a slightly lower V_{oc} than that of pure Pt CEs. The V_{oc} of DSSC depended on the discrepancy between the quasi-Fermi level of the electrons in the TiO₂ layer and the oxidation and reduction potentials of electrolyte. In this case, DSSCs were constructed by identical photoanode structures and electrolyte but with various CEs. Therefore, the different V_{oc} can be ascribed to the slightly poor electrocatalytic activity of PProDor-Bz₂/Pt composites CEs. The higher PProDor-Bz₂ concentration of CEs surface exhibited lower Voc (Bora et al. 2015). However, from Table 1, it could be also demonstrated that the PProDor-Bz₂/Pt composites CEs had higher J_{sc} than that of Pt CEs, which is due to its porous structures. The 25 mC cm⁻² PProDOT-Bz₂/10 s Pt and 25 mC cm⁻² PProDOT- Bz₂/30 s Pt composite counter electrodes provided the η of 3.62% and 4.72%, which were higher than those of 10 s Pt (3.20%) and 30 s Pt (3.77%) CE. The η for them increase 13% and 25%, respectively.



Fig. 1 (a) Schematic procedures of the PProDOT-Bz₂/Pt composite counter electrodes and (b) schematic diagram of DSSCs structure

CEs	V _{oc} (V)	$J_{sc} (mA/cm^2)$	FF	η(%)	Ref.
10s Pt	0.70	6.53	0.70	3.20	In this study
30s Pt	0.66	7.79	0.73	3.77	In this study
25 mC cm ⁻² PProDOTBz ₂ /10 s Pt	0.62	9.17	0.64	3.62	In this study
25 mC cm ⁻² PProDOTBz ₂ /30 s Pt	0.62	11.81	0.65	4.72	In this study
50 mC cm ⁻² PProDOTBz ₂ /10 s Pt	0.60	8.68	0.64	3.33	In this study
50 mC cm ⁻² PProDOTBz ₂ /30 s Pt	0.60	8.38	0.67	3.42	In this study
Pt	0.72	9.50	0.65	4.40	Xia et al. 2011
PPy	0.68	9.20	0.52	3.40	Xia et al. 2011

Table 1 The photovoltaic parameters of the DSSCs with different CEs



Fig. 2 The J-V curves of DSSCs based on different counter electrodes

3.2 Electrochemical impedance spectra

Figure 3 shows the Nyquist plots of the DSSCs based on various counter electrodes. The values of RS, R_1 and R_2 obtained by fitting the Nyquist plots are summarized in Table 2. The RS value could be estimated using the onset point of the first semicircle in the high frequency zone. A lower RS presents a better adherence of the catalytic layer onto the substrate and thereby gives rise to a good electrical conductivity of the film and good fill factor (FF) of DSSC (Chen *et al.* 2015). The first

semicircle in the high frequency region of EIS indicates the charge transfer resistance (R_1) at the interface between electrolyte and composite CE, which determines electrocatalytic performances of the counter electrode towards $I3^-$ reduction (Bora *et al.* 2015). The second semicircle indicates the charge transfer resistance at the interface of the photoanode and electrolyte (R_2) (Mehmood 2016). The CE based on PProDOT-Bz₂/Pt showed a lower R_1 value than that of Pt CE.



Fig. 3 The Nyquist plots of the DSSCs based on various counter electrodes

Table 2	The capacitance an	d resistance of the	e equivalent	circuit for	DSSCs able
	1				

	CEs	Rs (Ω)	$C_1 (\mu F)$	$R_{1}(\Omega)$	$C_2 (mF)$	$R_{2}\left(\Omega ight)$
	10s Pt	54.86	25.82	12.51	0.61	65.03
	30s Pt	27.67	82.36	2.98	0.79	23.01
25 r	nC cm ⁻² PProDOTBz ₂ /10 s Pt	29.54	47.30	6.76	0.97	36.81
25 r	nC cm ⁻² PProDOTBz ₂ /30 s Pt	22.19	114.97	2.57	1.77	21.96
50 r	nC cm ⁻² PProDOTBz ₂ /10 s Pt	32.33	71.23	5.91	0.93	27.72
50 r	nC cm ⁻² PProDOTBz ₂ /30 s Pt	27.52	53.71	3.96	1.08	23.74

3.3 Ehe photovoltaic parameters of the DSSCs with 25 mC cm⁻² PProDOT-Bz₂/Pt composite counter electrodes at different light intensities

Figure 4 displays the J–V curves of DSSC based on 25 mC cm⁻² PProDOT-Bz₂/30 s Pt counter electrode at different light intensities. The corresponding photovoltaic parameters are summarized in Table 3. The 25 mC cm⁻² PProDOT-Bz₂/30 s Pt composite counter electrodes at 100 mW cm⁻², 80 mW cm⁻², 50 mW cm⁻², 30 mW cm⁻² and 10 mW cm⁻² provided the η of 4.72%, 4.85%, 5.38%, 5.73% and 3.70%, respectively. However, the η of PProDOT-Bz₂/30 s Pt CE increased with decreasing the light intensity. The improvement in efficiency can be attributed to less diffusion overpotential and less electron recombination of the cell under 80 mW cm⁻², 50 mW cm⁻² and 30 mW cm⁻² than that of 100 mW cm⁻² (Lan *et al.* 2012). Moreover, Fig. 5 shows the simulated solar light source spectrum.



Fig. 4 The J-V curves of the DSSC based on 25 mC cm⁻² PProDOT-Bz₂/30 s Pt counter electrode at different light intensities

Table 3The photovoltaic properties of the DSSC with 25 mCcm⁻²PProDOT-Bz₂/30 sPtcounter electrode atdifferent light intensities

Light intensity (mW cm ⁻²)	V _{oc} (V)	J _{sc} (mA/cm ²)	FF	η(%)
100	0.62	11.81	0.65	4.72
80	0.62	9.27	0.68	4.85
50	0.62	5.99	0.73	5.38
30	0.62	3.74	0.74	5.73
10	0.54	0.88	0.78	3.70



Fig. 5 The spectrum of solar simulator.

4. CONCLUSIONS

This study investigated the impact of various Pt sputtering time of PProDOT-Bz₂/Pt composite counter electrodes and various illumination intensities in DSSCs. The DSSC based on PProDOT-Bz₂/10 s Pt composite counter electrode exhibited a PCE of 3.62%, which is compared with the 30 s Pt CE (3.77%) under the 100 mW cm⁻² light illumination. The DSSC based on 25 mC \mbox{cm}^{-2} PProDOT-Bz_2/30 s Pt composite films gave the photovoltaic conversion efficiency of 4.72% and 5.73%, under the 100 mW cm⁻² and 30 mW cm⁻² light intensities, respectively. From EIS analysis, the PProDOT-Bz₂/Pt composite CEs with smaller charge transfer resistances caused higher Jsc and η . Accordingly, the PProDOT-Bz₂/Pt composite CEs are deemed as promising candidates to replace Pt as an inexpensive counter electrodes material in DSSCs. Because the DSSC has better photovoltaic performances under low light intensity, it is important to further investigate the cell performance under low illumination intensity.

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