



A light-scattering co-adsorbent for performance improvement of dye-sensitized solar cells



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ABSTRACT

A light-scattering cyanobiphenyl derivative 6-[(4'-cyano(1,1'-biphenyl)-4-yl)oxy]hexanoic acid (CBHA) was designed, synthesized and applied as a co-adsorbent for dye-sensitized solar cells. The effect of CBHA on the light absorption and reflectance of dyed-TiO₂ photoanodes was investigated in detail. It revealed that a certain amount of CBHA adding to dye Z907 could obviously increase the light absorption of TiO₂ photoanodes by decreasing the dye loading amount and possibility of aggregation. Meanwhile, light-scattering property of CBHA rendered their TiO₂ photoanodes higher light-harvesting efficiency. In addition, electrochemical impedance spectroscopy results indicated that CBHA could restrain the interfacial electron recombination for longer life time. Thus, a performance improvement of ~0.81% from 5.23% to 6.04% could be obtained by the resulting device employing CBHA. Moreover, the DSSCs with CBHA also display better stability than the referenced devices. This novel light-scattering co-adsorbent offers us a feasible strategy to design multiple functional co-adsorbents for high performance dye-sensitized solar cells.

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1. Introduction

Dye-sensitized solar cells (DSSCs) are considered to be one of the most promising alternatives to conventional silicon-based photovoltaic devices due to their easy fabrication, flexibility, low cost and high solar energy conversion efficiency [1–6]. Recently, Mathew and co-workers employed SM315 dye with donor- π -bridge-acceptor and a Co (II/III) tris(bipyridyl) redox shuttle to manufacture the state-of-the-art devices, and the energy conversion efficiency exceeded 13% [7]. Although so high efficiency has been received, considerable efforts are still desirable to enhance the performance of the devices.

As we know, a typical DSSC comprises a nano-crystalline TiO₂ film deposited on a transparent conducting oxide glass, a Pt counter-electrode, and an electrolyte solution with a hole-transport mediator such as an iodide/triiodide redox couple, among which TiO₂ film is one of the core compositions. In general, charge separation and recombination often occur simultaneously at the TiO₂/dye/electrolyte interface. It has been reported that high

specific surface of TiO₂ photoanodes offer relatively big interface and enough electron injection [8]. Generally, according to most of previous reports, a surface area from 50 to 100 m²g⁻¹ is very suitable to ensure enough dye absorption. However, larger value of mesoporous TiO₂ particles will form much more surface interfaces, increasing the possibility of charge recombination between TiO₂ particles and I₃⁻ in the electrolyte. Meanwhile, larger value of mesoporous TiO₂ particles indicates much more grain boundaries among the particles, which also restrain the electron transport in the TiO₂ photoanode and ultimately decreases the device efficiency. Several strategies referring to the modification of the TiO₂ surface have been adopted to solve the above problems. For example, SnO₂ [9], Al₂O₃ [10] and Nb₂O₅ [11] are employed to deposit a thin inorganic insulating layer on the surface of the TiO₂ particles to inhibit the recombination of the photo injected electrons with their counter holes. Nevertheless, a too thick insulating layer maybe reduces the charge injection efficiency, ultimately resulting in lowering the energy conversion efficiency. Therefore, small organic molecules have attracted much attention utilized as co-adsorbents onto TiO₂ surface in DSSCs [3,6]. The employment of co-adsorbents can avoid the aggregation of dye molecules and reduce unfavourable energy transfer and quenching of photo-excited states [12,13]. In addition, the combination of dyes and co-adsorbents can form a more compact layer than do dyes

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alone, [14] which can effectively restrain the combination of electrons with the oxidized species (e.g. I_3^-). Furthermore, in some cases, they will also lead to more light-adsorption in UV and/or visible area, thus, enhance the light-harvesting efficiency of TiO_2 photoanodes for better device performance [7,11,15]. Phosphonic acid [16,17], bases [18], carboxylic acids [8,14] and acetyl acetone-typed compound [19] as appropriate adsorbents, have been proven to be effective to improve photovoltaic performance of DSSCs.

As known, cyanobiphenyl derivatives are a class of optical materials with excellent light-scattering properties [20,21]. For some compounds, they perform strong diffused reflectance during the visible light probably because of some unique molecular orientation. The rigid cyanobiphenyl groups always render these mesogens to form lamellar or columnar or reorientational macrostructures with light-scattering ability [22–24]. This triggered our interest in exploring the possibility of light-scattering to develop simple and new co-adsorbents with cyanobiphenyl structure for DSSCs. Herein, we successfully designed and synthesized a light-scattering co-adsorbent, 6-[(4'-cyano(1,1'-biphenyl)-4-yl)oxy]hexanoic acid, denoted as CBHA, which could protect the dyes adsorbed on the surface of TiO_2 from aggregation. Meanwhile, it showed obvious light-scattering property, leading to enhancement of light re-absorption by dyed photoanodes. Furthermore, the molecule dimension of CBHA was smaller than the dye molecule, making it suitable for compact monolayer formation through covering the bare TiO_2 surface during the dye Z907 uptake process. It has been found that CBHA manifested excellent performance when further applied as a co-adsorbent for DSSCs.

2. Experimental

2.1. Materials

18-crown-6, 4'-cyano-4-hydroxybiphenyl, methyl-6-bromohexanoate, and 1,2-dimethyl-3-propylimidazolium (DMPII), 4-tert-butylpyridine (TBP), guanidinium thiocyanate (GSCN), Chenodeoxycholic Acid (CDCA) were purchased from Alfa Aesar and used as received. Potassium carbonate was of analytical-reagent grade and purchased from China National Pharmaceutical Group Corporation. $TiCl_4$, H_2PtCl_6 and *cis*- $RuLL'(SCN)_2$ ($L = 4,4'$ -dicarboxylic acid-2,2'-bipyridine, $L' = 4,4'$ -dinonyl-2,2'-bipyridine) (dye Z907) were purchased from Aldrich, and all the chemical reagents were used without further treatment. Fluorine-doped tin oxide (FTO) glass electrodes ($8 \Omega/Sq$), and slurries containing 20 nm-sized mesoporous and 200 nm-diameter light-scattering TiO_2 colloidal were purchased from Dalian Hepat Chroma Solar Tech. Co., Ltd (China).

2.2. Synthesis of CBHA

The synthesis process of CBHA in detail is described as reported (in ESI†) [25]. 1H NMR (400 MHz, d_6 -DMSO): 7.85 (s, 4H), 7.69 (m, 2H), 7.04 (s, 2H), 4.00 (s, 2H), 2.51 (s, 2H), 2.22 (s, 2H), 1.72 (m, 2H), 1.56 (m, 2H), 1.43 (m, 2H). IR (KBr): 3410 cm^{-1} (O-H), 2939 cm^{-1} (H-C-H), 2228 cm^{-1} (C=N), 1703 cm^{-1} (C=O), 1252 cm^{-1} (C-C/C-O).

2.3. Device fabrication

The fabrication of DSSCs is represented as follows [26,27]. The FTO glass was cleaned by acetone and ethanol successively. Then, clean FTO glass was immersed into 40 mM aqueous $TiCl_4$ solution at 70°C for 30 min and washed with water and ethanol. Firstly, a $10\ \mu\text{m}$ -thick film of 20 nm-sized TiO_2 particles was coated onto the FTO glass by the doctor-blade technique and then dried at 125°C for 5 min. Secondly, a $5\ \mu\text{m}$ -thick layer of 200 nm light-scattering

TiO_2 particles was deposited on the top of the first TiO_2 layer. The two layers of TiO_2 particles were used as photo anode. The resulting TiO_2 films were annealed at 500°C for 30 min and gradually cooled to 90°C . The obtained TiO_2 electrode was soaked in a Z907 solution (0.5 mM) containing CBHA with molar ratios of 1:0, 2:1, 1:1, 1:2 in acetonitrile/tert-butyl alcohol (1: 1, v: v) for 24 h. Afterward, the dye-sensitized TiO_2 electrode was washed with anhydrous ethanol and dried with a N_2 stream. To prepare the Pt counter electrode, one drop of 5 mM H_2PtCl_6 in ethanol was dipped onto the cleaned FTO glass substrate, followed by dried and annealed at 400°C for 15 min. A typical liquid electrolyte was prepared by adding 1 M DMPII, 0.1 M I_2 , 0.5 M TBP and 0.1 M GuSCN in 3-methoxypropionitrile. The TiO_2 electrodes and platinized conducting glass were separated by using a $25\ \mu\text{m}$ thick Surllyn hot-melt ring (DuPont) and sealed up by heating. The liquid electrolyte was injected into sandwiched cells using a vacuum back filling system. The hole for electrolyte injection on the platinized FTO counter electrode was finally sealed with a Surllyn sheet and a thin glass by heating.

2.4. Characterization and Measurements

1H NMR spectra were recorded on a Varian 400 MHz spectrometer. Fourier transform infrared (FTIR) spectra of the synthesized compounds were recorded on a Varian CP-3800 spectrometer in the range of $4000\text{--}400\text{ cm}^{-1}$. Thermal analysis was carried out on Universal Analysis 2000 thermo gravimetric analyzer (TGA). Sample was heated from 50 to 500°C at a heating rate of $10^\circ\text{C min}^{-1}$ under N_2 flow. Differential scanning calorimetry (DSC) measurement was performed under N_2 atmosphere with a heating rate of $10^\circ\text{C min}^{-1}$ in a temperature range of $-50\text{--}250^\circ\text{C}$ on DSC-Q200. Ultraviolet-visible (UV-vis) absorption spectra and diffuse reflectance spectra with an integrating sphere were obtained using Shimadzu UV-2450 spectrophotometer. To obtain the relative values of the dye load, the dye Z907 was desorbed from the TiO_2 surface using 10 mM KOH ethanol solution. The solution was tested by the UV-vis absorption spectrum. Meanwhile, a series of standard ethanol solution containing different amounts of Z907 was prepared, and tested by their absorption spectrum with different intensity at 521 nm for a lineal relationship. According to the classical Lambert-Beer Law ($A = \epsilon cl$, where A is the measured absorbance, ϵ is absorptivity coefficient, c is the analyte concentration, and l is the path length), one could obtain the dye surface excesses in a unit of mol/cm^2 by comparing with the amounts (g) of TiO_2 particles on the FTO film, the molecular weight of Z907 (870.10), and the surface area $\sim 56\text{ cm}^2/\text{g}$ of TiO_2 particles used. Meanwhile, one could also obtain the probable amount of CBHA on the TiO_2 surface by recording the UV absorption intensity of CBHA at 298 nm and calculating the data. The photocurrent density-voltage (J-V) curves of the assembled DSSCs shielded by an Al foil mask with an aperture area of $\sim 0.25\text{ cm}^2$ were measured with a digital source meter (Keithley, model 2612) under simulated air mass (AM) 1.5 solar spectrum illumination at 100 mW cm^{-2} . The irradiation intensity has been calibrated by a Newport standard silicon solar 91150. Newport monochromator 74125 and power meter 1918 with silicon detector 918D are also used in the IPCE measurements at presented wavelength from 300 to 850 nm. The electrochemical impedance spectra (EIS) of the devices were acquired on a CHI660c electrochemical workstation using the AC impedance method at the forward bias voltage of -0.70 V and the frequency of $0.01\text{--}10^5\text{ Hz}$ under dark conditions and the amplitude is 5 mV. The equivalent circuit model for EIS analysis was used by Autolab (PGSTAT30). Incident photocurrent conversion efficiency (IPCE) plotted as a function of excitation wavelength was recorded on a Keithley 2612 source meter under the irradiation of a Xenon lamp with a monochromator (Oriel

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