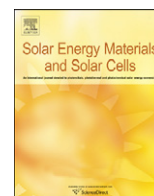




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Surface passivation: The effects of CDCA co-adsorbent and dye bath solvent on the durability of dye-sensitized solar cells

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ABSTRACT

The long-term stability of dye-sensitized solar cells (DSCs) based on an amphiphilic ruthenium sensitizer, CYC-B6L was studied. When the surface of the TiO₂ electrode was passivated with a co-adsorbent, chenodeoxycholic acid (CDCA), the resulting DSC reveals very good long-term stability with over 87% of initial performance under a 1000 h prolonged light-irradiation and thermal stress (60 °C) aging, which is as good as the art robust device. The solvent used in dye bath also affects the long-term stability of the DSC devices. The effect of co-adsorbent and the impact of the dye bath solvents on the device durability were investigated using the electrochemical impedance spectroscopy.

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1. Introduction

Dye-sensitized solar cells (DSCs) have become an alternative to the photovoltaic technology for converting sunlight energy into electricity, because of their low manufacture cost, tunable absorption range, and impressive conversion efficiencies [1]. DSCs consist of a nano-crystalline TiO₂ porous film, a monolayer of self-assembled dye (sensitizer), and a hole-transport mediator such as an iodide/triiodide redox couple. The photon-to-current conversion is achieved by injecting electrons from the photo-excited dye molecules into the conduction band of TiO₂. The hole in the dye is regenerated by oxidizing the iodide to iodine. The charge diffuses from TiO₂ semiconductor to the electron collector consequently reduces the oxidized mediator *via* an external circuit and counter electrode [2–4]. DSCs sensitized by ruthenium sensitizers in combination with the volatile electrolytes have achieved remarkable efficiencies of over 11% [5–9]. However, the stability of such highly efficient devices is poor, because of the leakage of the volatile acetonitrile-based electrolyte. Attempts to improve the durability of DSCs to reach the practical applications including the enhancement of the electrolyte robustness [10–24] and anchoring stability [25–26] of the sensitizers [7–9,16,27–33],

as well as the passivation of the surface of the porous TiO₂ film [34–42].

Amongst the available co-adsorbents for DSCs, chenodeoxycholic acid [34–35] (abbreviated CDCA, Fig. 1) has found to be extensively used for the devices sensitized with ruthenium complexes [7–9], organic dyes, and so on [43–54]. The dye molecules adsorbed on the porous TiO₂ film decrease as the CDCA concentration in the dye bath increases. This is because of the sensitizer and co-adsorbent self-assembled on the restricted TiO₂ surface is a competitive process although an improvement of the cell performance was observed by using a co-adsorbent. The improvement of the photovoltaic performance has been explained by three processes: (1) CDCA may fill the vacancy of the dye-adsorbed TiO₂ film so that the back current (from the charge recombination of the electron in mesoscopic TiO₂ film to the electrolyte) is reduced [34]. (2) Increases the Fermi level of TiO₂ film (shifts to more negative potential) [35]. (3) The presence of CDCA reduces the dye aggregation, facilitating the charge separation [38,43]. We are interested in understanding the correlation between the device durability and the amount of CDCA adsorbed on TiO₂ film.

In this paper, we report the long-term stability of the DSC devices based on an amphiphilic ruthenium super-sensitizer, coded CYC-B6L [55] (Fig. 1), fabricated in the presence of various amounts of CDCA in the dye bath. We used the electrochemical impedance spectroscopy (EIS) [56–65] to study the variations of the electron transport resistance (R_w) of the TiO₂ photo anode, the charge-transfer resistance (R_k) related to the electron

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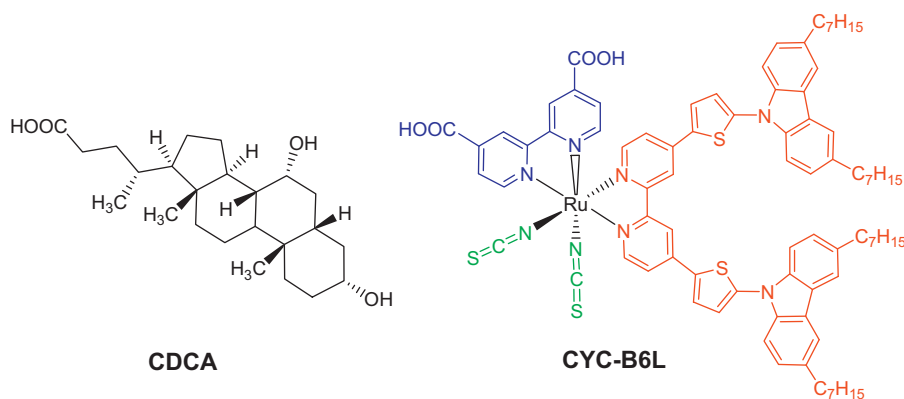


Fig. 1. Molecular structures of CDCA and CYC-B6L.

combination, effective electron lifetime (τ_{eff}) as well as the electron diffusion coefficient (D_{eff}) and diffusion length (L_n) of the devices during the light-irradiation and thermal aging processes. Furthermore, several solvents were used to prepare the dye solutions to investigate the effects of solvent in the dye bath on the long-term stability of the DSC device.

2. Experimental section

2.1. Materials

CYC-B6L ruthenium dye was prepared according to the paper published previously [55]. Chenodeoxycholic acid (CDCA, Sigma-Aldrich), *L*-propyl-3-methylimidazolium iodide (PMII, Merck), anhydrous I₂ (Sigma-Aldrich), *N*-methylbenzimidazole (NMBI, Sigma-Aldrich), 3-methoxypropionitrile (MPN, Alfa Aesar), acetonitrile (AN, HPLC grade, J.T. Baker), 4-*tert*-butylpyridine (TBP, Sigma-Aldrich), *t*-butanol (*t*-BuOH), dimethylformamide (DMF), tetrahydrofuran (THF), terpineol (anhydrous, #86480, Fluka), and ethyl cellulose (EC, Fluka) were used as received.

2.2. DSC device fabrication

Anatase TiO₂ nanoparticles (NP) (20 nm diameter) were hydrothermally prepared in a Ti-based autoclave as reported previously [66]. The screen-printable 20 nm TiO₂ paste was prepared by thoroughly mixing of 4.23 g TiO₂ NP, 55 ml ethyl cellulose, and 45 ml terpineol. An approximately 18 μm thick TiO₂ film was deposited on a 2.2 mm thick fluorine-doped tin oxide glass (FTO glass, Pilkington, TEC-8, 8 Ω/square) pre-treated with TiCl₄ by a screen-printing procedure. The screen-printed TiO₂ electrodes were annealed at 490 °C for 1 h in a stream of air, followed by immersion in the 40 mM TiCl₄ aqueous solution at 70 °C for 30 min. The electrodes were washed with ethanol and then water before being annealed further at 500 °C for 30 min. After cooling to the room temperature, the TiO₂ electrodes were immersed in a CYC-B6L dye bath at 60 °C for 12 h. The dye solutions were prepared by dissolving 0.3 mM CYC-B6L in a mixture of *tert*-butyl alcohol (*t*-BuOH) and AN ($v/v=1/1$), with different concentrations of CDCA co-adsorbent (0 mM, 1 mM, 5 mM and 10 mM for devices A, B, C and D, respectively). For devices E, F, G and H, additional 4 μm scattering layer (composed of 400 nm TiO₂ paste made by thoroughly mixing of 4.23 g mixture of 20 nm and 400 nm diameter TiO₂ ($w/w=1/1$), 55 ml ethyl cellulose, and 45 ml terpineol) was deposited on TiO₂ electrodes. The annealing and post-treatment processes for TiO₂ film with scattering layer are the same as those described above. The dye solution, consisting of 0.3 mM CYC-B6L and 5 mM of

CDCA, was dissolved in a mixture of *t*-BuOH and AN ($v/v=1/1$) (for device E), a mixture of TBP and AN ($v/v=1/1$) (for device F), DMF (for device G) and THF (for device H), respectively. The dye adsorbed TiO₂ working electrodes were rinsed with AN, and then dried at 60 °C for 5 min. Thermally platinized FTO glass was used as the counter electrode and a low-volatility electrolyte with the composition of 0.8 M PMII, 0.1 M I₂, and 0.5 M NMBI in MPN was used for all DSC devices. The hermetically sealed cells were wrapped with a 50 μm-thick polyester film (Preservation Equipment Ltd, UK) as an UV (below 400 nm) cutoff filter. The active area of the working electrode was 0.28 cm². The amount of CYC-B6L adsorbed on the 18-μm thick TiO₂ film was calculated by desorbing the dye with a solvent composed of TBP and tetrabutylammonium iodide (TBAI) ($v/v=1/1$) and measured with an UV/Vis. spectrometer.

2.3. Durability, photoelectrochemical, and electrochemical impedance spectroscopy (EIS) studies

The visible light irradiation (100 mW/cm²) and thermal stress (60 °C) aging of the hermetically sealed cells was performed with a Sun test xenon arc lamp (ATLAS Ci3000 xenon Fadeometer). Photoelectrochemical measurements were carried out at open circuit conditions after a period of continuous light irradiation, being allowed to cool and equilibrate at room temperature. We used an AM 1.5 G solar simulator (Yamashita Denso, YSS-100A) as the irradiation source for the current density–voltage ($J-V$) and electrochemical impedance spectroscopy (EIS) measurements. The intensity of the simulated sunlight was calibrated to be 100 mW/cm² by a silicon photodiode. Related data were collected by an electrochemical analyzer (Autolab, PGSTAT30) at 25 °C. For EIS measurements, a forward bias set at the open circuit voltage (V_{oc}) with AC amplitude of 10 mV between the working and counter electrodes was applied and the measured frequency was ranged from 50 MHz to 100 mHz. The equivalent circuit model for EIS analysis was similar to that used in the previous reports [67–69].

3. Results and discussion

3.1. The effects of CDCA on the device durability

The self-assemblies of the dye and co-adsorbent on the finite surface area of a porous TiO₂ film are competitive. DSC devices were fabricated by dipping the TiO₂ electrodes in the CYC-B6L dye baths with various CDCA concentrations (0 mM, 1 mM, 5 mM and 10 mM for devices A, B, C and D, respectively) for depositing different amount of dyes and co-adsorbents. Assuming that

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