



Dye mixture promoted light harvesting for organic dye-sensitized solar cells using triphenylamine dyes with various numbers of anchoring groups



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ABSTRACT

Co-sensitizers and co-adsorbents are promising materials to enhance the light harvesting efficiency and reduce the un-expected back transfer reaction (recombination) of dye-sensitized solar cells (DSSCs). In this study, three sensitizers with triphenylamine as an electron donor, thiophene as a bridge and various numbers of acceptors/anchors cyanoacetic acid (TPA3T1A, TPA3T2A and TPA3T3A) were synthesized, and TPA3T1A and TPA3T2A were used as co-adsorbents with TPA3T3A. The results showed that co-adsorption on the TiO₂ surface at the following percentages, TPA3T3A 73%, TPA3T1A 17% and TPA3T2A 10%, resulted in an increase in the photovoltaic performance of the DSSCs from 5.27% to 5.83% compared to that of a single TPA3T3A sensitizer due to the increasing J_{SC} and V_{OC}. This enhancement might be due to improved light absorption and decreasing recombination by the co-sensitizers, TPA3T1A and TPA3T2A, occupying all the empty places on the TPA3T3A-adsorbed TiO₂ surface.

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1. Introduction

Solar energy is one solution for alternative global energy sources because of its unlimited, environmentally-friendly and renewable properties. Among photo-electrochemical solar cells, dye-sensitized solar cells (DSSCs), which were first reported in 1991 by Professor Michael Gratzel, have attracted attention because they employ inexpensive materials and convenient manufacturing [1]. When DSSCs absorb sunlight, the electrons are generated from excited-photosensitizers and are then injected into a wideband gap semiconductor (TiO₂) and reach the counter electrode to form an external circuit. The photosensitizers are restored to their ground state by electron transfer from the redox electrolyte, normally an organic solvent containing the iodide/triiodide redox system [2–5].

An increase in the light harvesting efficiency of solar cells leads to an increase in current density due to the enhanced generated-electrons from photosensitizers. Light harvesting can be enhanced by the increased light reflection [6–9], Forster resonance energy transfer [10], broaden the light absorption band of the co-sensitizer [11–15]. A back transfer reaction (recombination)

between the injected electron in TiO₂ and the electrolyte is one of the un-expected reactions in DSSCs because it causes a loss of potential. Therefore, reduced recombination may increase the electron lifetime and improve the open-circuit voltage of DSSCs, leading to an increase in the photovoltaic performance. Increasing the bulky structure of sensitizers or redox complexes [16], using a co-adsorbent as a blocking layer [17–21] could insulate the TiO₂ surface to limit the approach of the oxidized electrolyte and injected electron in TiO₂, leading to a decrease in the recombination rate.

Three triphenylamine-based sensitizers with different numbers of acceptors/anchors TPA3T1A, TPA3T2A and TPA3T3A were synthesized based on the donor-linker-acceptor (D- π -A) structure (Fig. 1). With increasing number of acceptors/anchors, the molar extinction coefficient and force adsorbed, electronic coupling of the sensitizer and TiO₂ were enhanced [22] resulting improved photovoltaic performance, and TPA3T3A produced the highest performance compared to TPA3T1A and TPA3T2A. From these results, an attempt was made to improve the efficiency of DSSCs using TPA3T3A as a sensitizer by filling the empty places on the TPA3T3A-adsorbed TiO₂ with TPA3T1A and TPA3T2A. The sensitizers, TPA3T1A and TPA3T2A, may not only work as a co-sensitizer to improve the light harvesting efficiency, but also work as a blocking layer to reduce the recombination because the TiO₂ surface is

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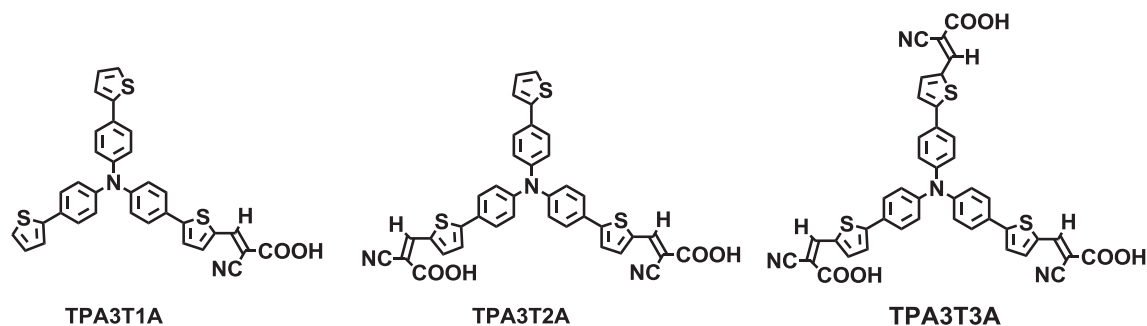


Fig. 1. Synthesis of TPA3T1A, TPA3T2A and TPA3T3A.

covered with sensitizers, which help improve the J_{SC} and V_{OC} , leading to the enhanced photovoltaic performance of DSSCs. Co-adsorption was conducted using the sensitizer solutions (0.3 mM) with TPA3T1A: TPA3T2A: TPA3T3A molar ratios of 1:1:1 and 1:1:5 (called solution 1:1:1 and solution 1:1:5) to compare with a solution of single TPA3T3A 0.3 mM and provide a deeper understanding of the co-adsorption effect on the photovoltaic performance of DSSCs.

2. Experimental section

2.1. Materials

All starting materials and solvents were purchased from Sigma Aldrich. Triphenylamine-based organic dyes, TPA3T1A, TPA3T2A and TPA3T3A, were prepared using the synthesis method reported elsewhere [22]. Fig. 1 shows the chemical structures.

2.2. Instruments

All ^1H NMR spectra were recorded on a Bruker Advance NMR 300 MHz spectrometer using $\text{DMSO-}d_6$. The UV–Vis spectra were measured using a Cary 5000 UV–Vis–NIR spectrophotometer. The photo-electrode thickness was measured using an Alpha-step IQ surface profiler (KLA Tencor).

2.3. Preparation of DSSCs

The fluorine-doped tin oxide (FTO) transparent conducting glasses (Pilkington, $15 \Omega/\text{cm}^2$) were cleaned ultrasonically with methanol, D.I water and acetone. A transparent meso-porous TiO_2 paste (20–30 nm in diameter, Dyesol Ltd.) was deposited on FTO glasses using the doctor blade technique, and sintered at 450°C for 30 min. The 200 nm in diameter TiO_2 (Dyesol Ltd.) as the scattering layer (200 nm in diameter, Dyesol Ltd.) was coated on transparent meso-porous TiO_2 films, followed by sintering at 450°C for 30 min. Subsequently, the TiO_2 films were immersed in a 40 mM TiCl_4 aqueous solution at 70°C for 30 min and sintered at 450°C for 30 min. After sintering, when the temperature was cooled to 70°C , the TiO_2 films were dipped in 0.3 mM dye solutions (TPA3T1A, TPA3T2A, TPA3T3A, solution of TPA3T1A:TPA3T2A:TPA3T3A at ratios of 1:1:1 and 1:1:5) at 25°C for 24 h in the dark and then rinsed with acetonitrile to remove the excess dye to obtain the photo-electrodes. The counter electrodes were prepared by coating a platinum paste on pre-drilled FTO glasses using the doctor blade technique, then heated to 450°C for 30 min. The photo-electrode and counter electrodes were combined with a 60 μm thick Surlyn film (Dupont) as a spacer. An electrolyte solution was then introduced through pre-drilled holes on the counter electrode. The

employed electrolyte was a solution of 1-butyl-3-methylimidazolium iodide (0.7 M), lithium iodide (LiI, 0.2 M), iodine (I_2 , 0.05 M), and 4-*tert*-butylpyridine (TBP, 0.5 M) in acetonitrile/valeronitrile (85:15, v/v).

2.4. Photovoltaic characterization

The photo current density-voltage (J-V) characteristics of the DSSCs were measured under irradiation with a solar simulator AM 1.5 with an incident light intensity of $100 \text{ mW}/\text{cm}^2$ (PEC-L11, Pecell Technologies, Inc.). The incident monochromatic photon-to-current efficiencies (IPCEs) were recorded as a function of the light wavelength using an IPCE measurement instrument (PEC-S20, Pecell Technologies, Inc.). Electrochemical impedance spectroscopy (EIS) was carried out using a computer-controlled potentiostat (IVIUMSTAT, IVIUM) at the open circuit voltage with a 10 mV of amplitude and an AC frequency range between 100 kHz and 0.1 Hz (PEC-L11, Pecell Technologies, Inc.). The electron diffusion coefficient and electron lifetime of the DSSCs were measured by the stepped light-induced transient measurements of the photocurrent and voltage (SLIM-PCV) using a same method reported by O'Regan et al. [23] and as illustrated elsewhere [24]: a white light bias was generated from an array of diodes (Abet, LS Series light source) with red-light pulsed diodes (Thorlabs HNL210L system) as the perturbation source, controlled by a fast solid-state switch. The voltage dynamics were measured on a 1 GHz Tektronix oscilloscope (DPO4102B-L) across a high impedance (1 M Ω) port. The perturbation light source was set to a suitably low level such that the voltage decay kinetics were mono-exponential. Small perturbation transient photocurrent measurements were conducted in a similar manner to the open-circuit voltage decay measurements. For the voltage decay measurements in the short-circuit regime, a Keithley 2600B source meter was connected in series to the solar cell and in parallel with the oscilloscope that was set on the high impedance port.

3. Results and discussion

Fig. 2 presents the absorption spectra of TiO_2 films (5 μm) after immersion into the TPA3T1A, TPA3T2A, TPA3T3A, 1:1:1, and 1:1:5 solutions, which absorbed incident light in the range, 350–600 nm, with a maximum absorption wavelength at approximately 445 nm. TPA3T3A shows the strongest light absorption compared to TPA3T1A and TPA3T2A. This might be due to the greater effect of the conjugated δ -chromophores with an increase in the number of acceptors/anchors [25,26]. The TiO_2 films dipped in a 1:1:1 solution exhibited higher light absorption compared to the single TPA3T1A and TPA3T2A but lower than TPA3T3A, whereas the TiO_2 film dipped in the dye solution 1:1:5 showed the highest absorption

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