

Triphenylamine-based tri-anchoring organic dye with enhanced electron lifetime and long-term stability for dye sensitized solar cells



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ABSTRACT

An organic sensitizer with a multi-anchoring system is a versatile methodology for enhancing the stability and conversion efficiency of dye-sensitized solar cells (DSSCs). Triphenylamine dyes (TPA3T1A~3A) containing different numbers of anchoring groups are synthesized to determine the correlation between the number of anchoring groups and photovoltaic properties as photosensitizers for DSSCs. The adsorption properties of the dyes on TiO₂ electrode were examined by ATR-FT-IR, which show that a mono-anchoring TPA3T1A system adsorbs in monodentate ester-type mode, and the three carboxylic acids in TPA3T3A adsorbs in bidentate bridging mode. The multi-anchoring dye exhibits strong electronic coupling with TiO₂, providing an efficient charge injection rate. Moreover, they increased the electron lifetime significantly by suppressing the charge recombination probability. This synergistic effect enables the fabrication of efficient photovoltaic devices. Furthermore, enhanced long-term stability is also observed in the DSSCs containing a tri-anchoring system compared to the mono- and bi-anchoring systems.

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1. Introduction

Dye sensitized solar cells (DSSCs) have attracted considerable attention as the next-generation of efficient solar cells [1–6], and have shown an overall peak power conversion of approximately 13%. In addition, they have been the focus of an economical solar electricity generation because the raw materials for producing DSSCs are relatively inexpensive, and the manufacturing process is rather simple.

The standard structure of a DSSC comprises of an electrochemical cell composed of a dye-adsorbed wide band gap semiconductor electrode, such as TiO₂ or ZnO, an electrolyte containing the I⁻/I₃⁻ redox couple, and a Pt-coated counter electrode [7–9]. The mechanism of DSSCs is based on the injection of electrons from the photosensitizer into the conduction band of nanocrystalline TiO₂ or ZnO. The oxidized photosensitizers are reduced by electron injection from the electrolyte. Therefore, the photosensitizer plays

an important role in capturing photons and generating electron/hole pairs, as well as transferring them to the interface of the semiconductor and electrolyte, respectively [5]. Ru-complexes are the most widely used photosensitizers for DSSCs because they exhibit high performance and good long-term stability [9,10]. They typically produce electricity with an 11.5% power conversion efficiency under 1 Sun conditions. Ru-complex dyes, however, contain the ruthenium metal ion, which is an expensive precious metal species that increases the cost of DSSCs. Therefore, the development of ruthenium-free organic dyes is essential for improving the commercialization of DSSCs.

A range of Ru-free organic photosensitizers have been evaluated as promising alternatives to Ru-complexes because of their many potential advantages [11–15]. Ru-free complexes are much cheaper and have higher molar extinction coefficients (e.g. the ϵ_{\max} of squaraine is $\sim 3 \times 10^5 \text{ M}^{-1} \text{ cm}^{-1}$) [16,17] than conventional dyes (e.g. the ϵ of Ru complexes is $\sim 1 \times 10^4 \text{ M}^{-1} \text{ cm}^{-1}$) [18]. They can also absorb photons with long wavelengths in the range, 500–700 nm. In addition, their synthetic process is convenient and their molecular design can be customized easily. Therefore, the development of novel Ru-free dyes for the production of high performance DSSCs will be a great challenge. Organic dyes,

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however, have limited applications, due mainly to their inferior energy conversion efficiency and stability compared to Ru-complexes dyes.

A novel strategy was recently introduced to the design of Ru-free organic dyes for the fabrication of efficient DSSCs [13]. Organic dyes containing multiple-electron acceptors/anchors are more efficient than single-electron acceptor/anchor types because the former provides more electron extraction paths between the photosensitizing dye and TiO₂, resulting in stronger electronic coupling with TiO₂. These types of DSSCs, however, are unstable compared to Ru-complexes because they can be desorbed easily from the TiO₂ surface in a liquid electrolyte.

In this study, a highly efficient photosensitizing system capable of operating with good stability was fabricated and optimized. In particular, a series of electron donor-acceptor (D-A) chromophores containing a triphenylamine unit as the donor and different numbers of cyanoacrylic acids as the anchors/acceptors with thiophene bridges in the chromophore for the DSSCs were designed and synthesized. The dyes contained a different number of anchoring moieties in their chromophores. Organic dyes containing multiple-electron acceptors/anchors are expected to be bound strongly to the Ti atoms on the surface of TiO₂, which improves the stability of the DSSCs significantly. The effect of the number of anchoring groups on the photovoltaic performance was investigated systematically by comparing their photophysical properties. Their photovoltaic performance was examined by measuring the photocurrent density-voltage (J-V) characteristics under irradiation with simulated solar light and the incident photon to current efficiency (IPCE). The interfacial electron transfer processes were analyzed by measuring the electrochemical impedance and open-circuit voltage decay (OCVD). The long-term stability of each DSSC was also examined by measuring the photovoltaic performance over a 1000 h period.

2. Experimental

2.1. Synthesis of organic dyes

Unless stated otherwise, all commercially available starting materials and solvents were purchased from Aldrich, TCI or Acros Co., and used as received. High performance liquid chromatography grade toluene and tetrahydrofuran (THF) were obtained from Samchun Chemical and distilled from CaH₂ immediately before use. Triphenylamine-based organic dyes, TPA3T1A, TPA3T2A and TPA3T3A, were prepared using the synthesis method reported elsewhere [19]. Fig. 1 shows the chemical structures.

2.2. Instrumental analyses

The ¹H-nuclear magnetic resonance (NMR, Advance NMR 300 MHz, Bruker) spectra were recorded using CDCl₃ and DMSO-*d*₆ solvents purchased from Cambridge Isotope Laboratories, Inc. The Fourier transform infrared (FT-IR, Perkin Elmer) spectra were obtained using a Miracle single bounce diamond ATR cell from PIKET Technologies.

2.3. Assembly and characterization of the DSSCs

The transparent conducting glass substrates (FTO; TEC8, Pilkington, 8 Ω/cm²) were cleaned in ethanol with ultrasonication. The TiO₂ pastes (particle size, ca. 20–30 nm in diameter) were prepared using ethyl cellulose (Aldrich), lauric acid (Fluka), and α-terpineol (Aldrich). Pre-cleaned glass substrates were coated with the prepared TiO₂ paste using the doctor blade method, followed by calcination at 500 °C for 30 min. The film thickness was measured using a surface profiler (Alpha-step IQ surface profiler, KLA Tencor). A scattering layer consisting of rutile TiO₂ particles (250 nm in a size) was deposited onto the mesoporous TiO₂ films. The layers were dipped into an aqueous solution of TiCl₄ (0.04 M) at 70 °C for 30 min. For dye adsorption, the TiO₂ electrodes were immersed into a dye solution (0.5 mM of dye in DMF) at 50 °C for 3 h. Pt counter electrodes were prepared by thermal reduction of the films dip-coated in H₂PtCl₆ (7.0 mM) in 2-propanol at 400 °C for 20 min. The dye-adsorbed TiO₂ and Pt counter electrodes were sandwiched between the 60 m-thick Surlyn (Dupont 1702), which was used as a bonding agent and spacer. A liquid electrolyte was introduced to the Pt counter electrode through a pre-punched hole and finally sealed. The electrolyte was composed of 3-propyl-1-methyl-imidazolium iodide (PMII, 0.7 M), lithium iodide (LiI, 0.2 M), iodine (I₂, 0.05 M), and *t*-butylpyridine (TBP, 0.5 M) in acetonitrile/valeronitrile (85:15). The active area of the dye-adsorbed TiO₂ films was estimated using a digital microscope camera using image-analysis-software (Moticam 1000).

The photovoltaic I–V characteristics of the prepared DSSCs were measured under a 1 sunlight intensity (100 mW cm⁻², AM1.5), which was confirmed using an AIST-calibrated Si-solar cell (PEC-L11, Peccell Technologies, Inc.). The incident monochromatic photon-to-current efficiencies (IPCEs) were plotted as a function of the wavelength of light using an IPCE measurement instrument (PEC-S20, Peccell Technologies, Inc.). The long-term stability was assessed under 1 Sun illumination at 30 °C for 1100 h. The impedance spectra (EIS) and open-circuit photovoltage decay (OCVD) curves were acquired using an electrochemical impedance analyzer (IVIUMSTAT, IVIUM). EIS is performed over the frequency range, 100 kHz and 100 MHz, with an AC voltage amplitude of

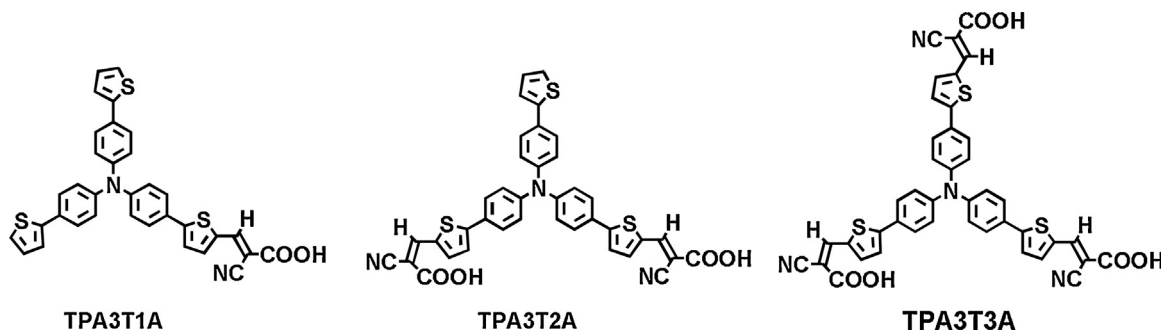


Fig. 1. Chemical structures of the synthesized organic dyes TPA3T1A, TPA3T2A, and TPA3T3A.

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