



Contents lists available at ScienceDirect

Journal of Photochemistry and Photobiology A: Chemistry

journal homepage: www.elsevier.com/locate/jphotochem

Disperse red 1 and reactive red 2 – modified nanoparticulate TiO₂ films as photoanodes for studying photovoltaic properties of textile azo dyes



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ARTICLE INFO

Article history:

Received 3 March 2015

Received in revised form 2 April 2015

Accepted 5 April 2015

Available online 9 April 2015

Keywords:

Dye-sensitized solar cells

Textile azo dyes

Charge carrier dynamics

ABSTRACT

Here we report the preparation of dye-sensitized solar cells containing disperse red 1 (**DR1**)- or reactive red 2 (**RR2**)-modified nanoparticulate TiO₂ photoanodes and their charge carrier dynamics under visible light illumination. Photoelectrochemical data obtained for both devices indicated that the photoinduced charge generation process was significantly better for **RR2**-sensitized solar cells than for **DR1**-sensitized solar cells. The effectiveness of the photoinduced charge generation observed for the **RR2**-sensitized TiO₂ photoanodes was explained by invoking two complementary phenomena which can be promoted in the presence of the voluminous 1,3,5-triazine moieties of **RR2**: (a) delay of the increment of I₃⁻ ions concentration into the dyed-TiO₂ pores, due to their known capabilities for establishing charge transfer complexes between N atoms of the heterocyclic ring and I atoms of the electrolyte; and (b) reduction of the electron interception process, due to the reaction established between photogenerated electrons and I₃⁻ anions of the electrolyte.

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1. Introduction

A large number of synthetic dyes have been employed for constructing dye-sensitized solar cells (DSSCs) containing nanoparticulate TiO₂ electrodes. Among others, ruthenium complexes and porphyrins have been successfully employed for assembling highly efficient DSSCs [1,2]. However, their high costs of preparation have motivated new investigations to identify technical grade sensitizers for constructing DSSCs of promising efficiencies.

Recently, metal-free organic sensitizers, including azo (—N=N—) dyes, have attracted attention because these molecules show significant molar absorptivity coefficients [3,4]. Particularly, textile azo dyes are commercially available compounds that have a lower cost than other metal-free organic dyes, motivating several investigations into their properties as photon receptors in DSSCs [3,5]. For instance, Chauhan et al. [3] prepared TiO₂ based DSSCs for studying light harvesting properties of fluorone, triarylmethane, thiazine and azo based commercial dyes as TiO₂ photosensitizers having sulfonic (SO₃⁻) and carboxylate (COO⁻) anchoring groups. Interestingly, the azo dyes – sensitized solar cells showed global

conversion efficiencies between 0.90 and 1.19%. On the other hand, Oprea et al. [5] reported a combined experimental and theoretical study for exploring the role of energy level alignment in solar cells sensitized with Mordant Yellow 10 (MY-10) having —SO₃H, —COOH and —OH anchoring groups. They found that the energy of the HOMO and LUMO levels were increased when the deprotonation of the sulfonic and carboxylic groups was carried out, thus promoting that the energy difference between HOMO and TiO₂ conduction band was bigger for enhancing the electron injection into the TiO₂ films. However, it was observed that the deprotonation of the hydroxyl groups causes the energy of the LUMO level that becomes higher than for the redox potential of the triiodide–iodide electrolyte, thus promoting that the dye regeneration process be thermodynamically unfavorable.

In this context, we propose that further investigations could be focused towards obtaining fundamental knowledge about the charge carrier dynamics in regenerative azo dye-sensitized solar cells, which should assist further studies into the limits of these devices as sources of electrical power. Here we prepared DSSCs containing nanoparticulate TiO₂ photoanodes by screen-printing deposition followed by sensitization with disperse red 1 (**DR1**, Fig. 1(A)) and reactive red 2 (**RR2**, Fig. 1(B)) to enable the study of the charge carrier dynamics in regenerative azo dye-sensitized solar cells.

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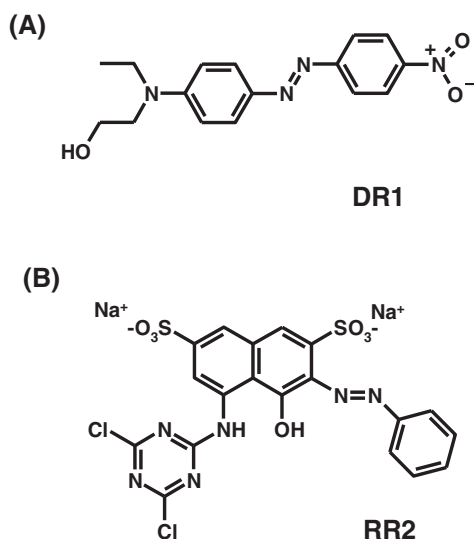


Fig. 1. Chemical structures for (A) disperse red 1 (**DR1**) and (B) reactive red 2 (**RR2**).

2. Experimental

2.1. Preparation of azo dye-sensitized solar cells

Nanoporous TiO_2 photoelectrodes ($5 \pm 1 \mu\text{m}$ -thickness estimated with a Dektak 6M Stylus Profiler (series of 5 samples) and

roughness factor of 523 ± 36 [6]) were prepared by following the doctor Blade technique, where a TiO_2 paste was screen-printed on optically transparent electrodes (OTEs, $2.5 \text{ cm} \times 1.5 \text{ cm} \times 2.2 \text{ mm}$, resistivity $\sim 15 \Omega \text{ sq}$, TEC15 Low-E Pilkington) having a F-doped tin oxide conductive layer [5]. TiO_2 paste was prepared by incremental addition of 0.5 mL of aqueous HCl 10% (v/v) to 300 mg of nanoparticulate TiO_2 powder (P25 Degussa, 21 nm-diameter, $50 \pm 15 \text{ m}^2/\text{g}$ specific area [7]) in a beaker while the mixture was homogeneously mixed with a glass rod. New TiO_2 layers were annealed in air at 450°C for 1 h prior to use [8]. Cathodes were prepared as follows: $8 \mu\text{L}/\text{cm}^2$ of a 10 mM $\text{H}_2\text{PtCl}_6 \cdot 6\text{H}_2\text{O}$ (Strem Chemicals, 99.9%-Pt) solution in 2-propanol (J.T. Baker, 99.97%) was homogeneously spread on the conductive face of OTEs. The new cathodes were subsequently dried at room temperature and then annealed in air at 380°C for 30 min [6].

TiO_2 electrodes were immersed for 24 h in MeCN (J.T. Baker, HPLC grade) solutions containing 3.18 mM **DR1** or 2.06 mM **RR2** dyes (Química Mexibras, Mexico, both technical grade) for sensitization at 25°C . Thereafter, dyed TiO_2 films were taken out from the dyeing solutions, rinsed with fresh MeCN, and immediately heated in a 360°C furnace for 1 h to eliminate residual MeCN. The resulting dye-modified electrodes were denoted as $\text{TiO}_2/\text{DR1}$ and $\text{TiO}_2/\text{RR2}$.

DSSCs were assembled in a sandwich fashion utilizing a $160 \mu\text{m}$ -thick o-ring (6 mm internal diameter; thickness measured with the same Dektak 6M Stylus Profiler), home-made from a CG3700 3M transparency sheet in order to avoid premature short circuits. An electrolyte solution of propylene carbonate (Aldrich, anhydrous, 99.7%) containing 0.3 M LiI (Aldrich, 99.9%) + 0.015 M I_2

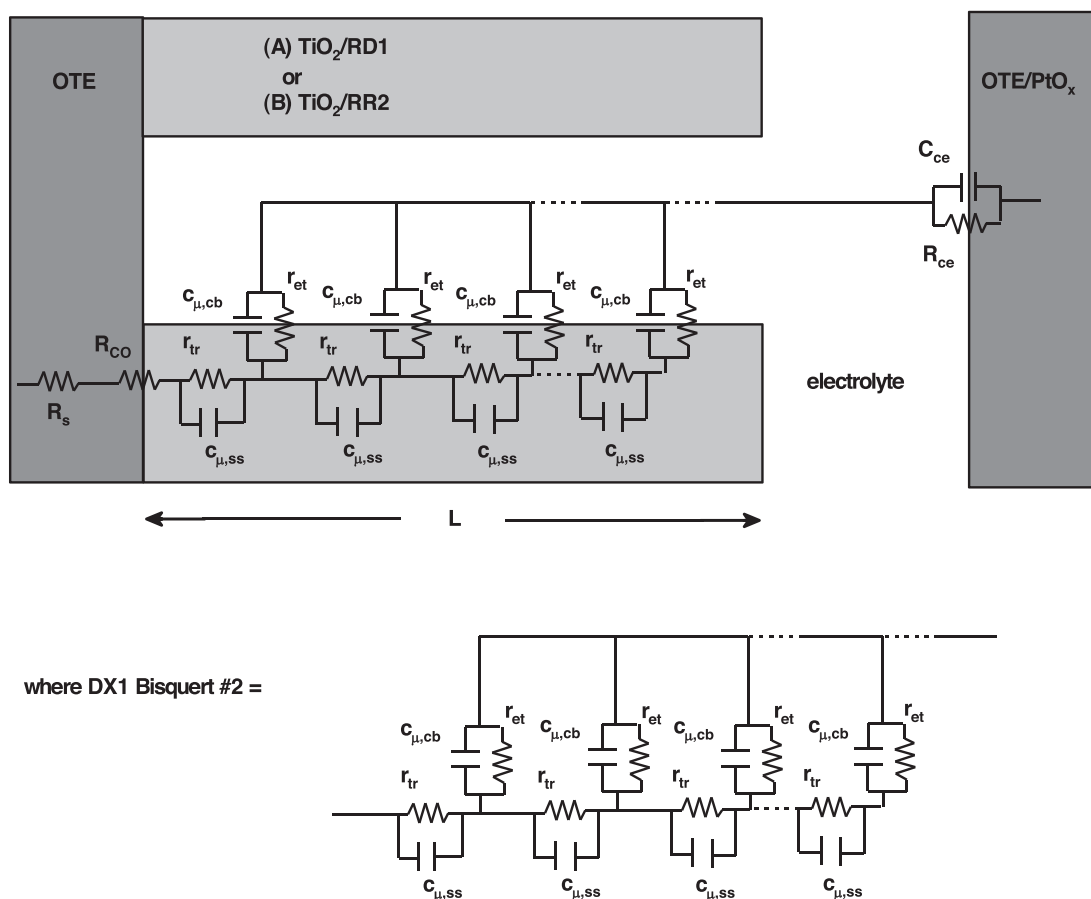


Fig. 2. Equivalent circuit fitted to the PEIS spectra experimentally obtained for DSSCs which contain: $\text{TiO}_2/\text{DR1}$ and $\text{TiO}_2/\text{RR2}$ electrodes.

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