



# Co-sensitization of N719 and RhCL dyes on carboxylic acid treated TiO<sub>2</sub> for enhancement of light harvesting and reduced recombination

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## ABSTRACT

We report herein the findings from a systematic study conducted on the effect of co-sensitization of two dyes i.e. N719 and rhodamine 19 perchlorate (RhCL) onto TiO<sub>2</sub> electrodes (thickness: 4 μm), which were treated by three different carboxylic acids namely, formic acid (FA), oxalic acid (OA) and citric acid (CA). Co-sensitization was carried out by first loading the carboxylic acid treated TiO<sub>2</sub> electrodes with RhCL dye followed by N719. The amount of dye loading was estimated from the UV/vis spectroscopy and the results show that: (i) the amount of individual dye loading (RhCL or N719) onto carboxylic acid treated TiO<sub>2</sub> electrodes is, in general, higher than that onto untreated TiO<sub>2</sub> electrodes; (ii) the amount of individual dye loading is highest for OA-treated and lowest for CA-treated TiO<sub>2</sub> electrodes; and (iii) the co-sensitization leads to highest loading of N719 onto FA-treated and lowest on CA-treated TiO<sub>2</sub> electrodes. The results of dye sensitized solar cells fabricated using these acid treated TiO<sub>2</sub> electrodes revealed that the efficiency ( $\eta$ ) is higher for electrodes having higher loading of N719 dye. For single N719 dye loading, the highest  $\eta$  of 4.5% is observed for OA-treated TiO<sub>2</sub>; while upon co-sensitization the highest  $\eta$  of ~5.3% is observed for FA treated TiO<sub>2</sub>. A detailed analyses of Fourier transform infrared spectroscopy (FTIR), UV–Visible, impedance, incident photon to current efficiency (IPCE) results show that  $\eta$  enhancement occurs due to the following factors: (i) increased short circuit current density ( $J_{sc}$ ) owing to high N719 dye loading which enhances light harvesting; (ii) improved IPCE; (iii) increased open circuit voltage ( $V_{oc}$ ) due to an upward shift of the conduction band edge (CBE) and quasi Fermi level; and (iv) suppressed back electron transfer.

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## 1. Introduction

Dye-sensitized solar cells (DSSCs) are increasingly being viewed with interest as potential devices for harnessing solar power. This is primarily on account of ease of fabrication and their relative low cost as compared to the

conventional silicon based solar cells [1–3]. A typical DSSC consists of dye sensitized mesoporous TiO<sub>2</sub> photoanode, electrolyte containing I<sub>3</sub><sup>-</sup>/I<sup>-</sup> redox couple and a platinum counter-electrode (CE). When the device is exposed to sunlight, the following processes occur: (i) the excited dye injects electrons into the conduction band of the TiO<sub>2</sub> electrode; (ii) these electrons move through nanocrystalline TiO<sub>2</sub> particles through external circuit towards the CE; (iii) the oxidized dye is regenerated via electron transfer from iodide ions; and (iv) the iodide ions are

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regenerated by the reduction of tri-iodide ions at the interface of CE and electrolyte [4]. The important photovoltaic parameters which govern the overall efficiency of the DSSCs are: (i) open circuit voltage ( $V_{OC}$ ), (ii) short circuit current ( $J_{SC}$ ) and (iii) fill factor (FF).  $V_{OC}$  is governed by the difference in the quasi-Fermi level of  $TiO_2$  electrode and the potential of the redox couple.  $J_{SC}$ , on the other hand, is primarily controlled by efficient injection of electron from the dye into conduction band (CB), amount of dye loading on  $TiO_2$  anode as well as charge collection at the counter-electrode. Therefore, the movement of the  $TiO_2$  conduction band edge (CBE) influences both  $V_{OC}$  and  $J_{SC}$ , which results in overall change in the device efficiency. The position of CBE of  $TiO_2$  depends on: (i) molecular interaction between dye molecules and  $TiO_2$  surface, (ii) electrolyte components and (iii) surface treatments of the  $TiO_2$  [5–9].

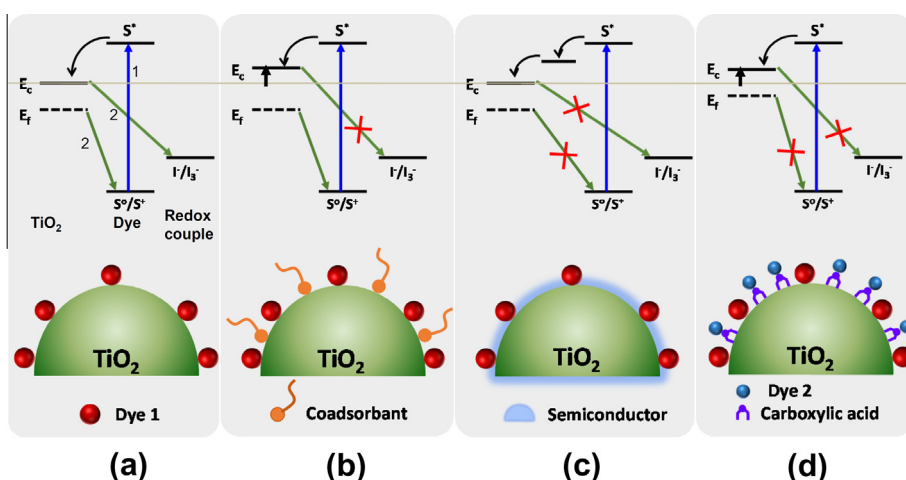
In addition to the position of CBE,  $V_{OC}$  and  $J_{SC}$  (and therefore, device efficiency) are also limited by the recombination processes of photo-injected electron with dye molecule and/or redox couple, as schematically demonstrated in Fig. 1a. In literature, many approaches, such as use of electrolyte additive, ferrocene/ferrocenium redox couple, surface treatment of  $TiO_2$  have been employed for retarding the recombination process [10]. Among these, surface modification of  $TiO_2$  is most adopted and different surface modification approaches adopted to reduce back electron transfer are as follows:

(i) Passivation of the surface states of  $TiO_2$  by co-adsorbents: In this approach, amphiphilic molecules containing carboxylic or phosphonic end groups, such as 4-guanidinobutyric acid, stearic acid, 3-phenylpropionic acid, are co-grafted on  $TiO_2$  surface along with dye, as shown schematically in Fig. 1b [11–13]. The carboxylic (or phosphonic) groups of the co-adsorbent passivate the surface states of

$TiO_2$  through binding to the unsaturated Ti, and thus, move the CBE upward with respect to the untreated  $TiO_2$  surface. The hydrophobic moiety acts as a shield between  $TiO_2$  CB electrons and redox species in the electrolyte which results in suppressed recombination and higher photo-voltage. Unfortunately, the co-adsorbent reduces the amount of dye loading on the  $TiO_2$  electrode. Nevertheless, this approach is useful in improving the device performance since the recombination rate reduction outweighs any decrease in dye loading.

(ii) Addition of a blocking/tunneling layer at  $TiO_2$  surface: In this case, as schematically shown in Fig. 1c, a blocking layer is first deposited onto the  $TiO_2$  electrode that acts as an energy barrier between CB of  $TiO_2$  and the redox potential of electrolyte. This blocking layer suppresses the recombination processes. The normally used blocking layers used are  $SiO_2$ ,  $Al_2O_3$ ,  $ZnO$ ,  $SrTiO_3$ ,  $Nb_2O_5$ ,  $SrO$  [14–22]. However, this approach requires a precise control on the thickness of the blocking layer as increased thickness can yield a decreased electron injection from the dye molecules to  $TiO_2$ , and therefore, a reduction in the  $J_{SC}$ . Nevertheless, this approach has been useful in improving the device performance since the recombination rate reduction is much more than the electron injection rate [17,20,21]. This approach, however, suffers from following two limitations: (a) the amount of dye loading decreases as blocking layer may reduce the number of anchoring sites of  $TiO_2$  surface for dye molecules, and (b) the CBE position remains almost unaffected, resulting in no improvement in  $V_{OC}$ .

(iii) Co-sensitization of formic acid treated  $TiO_2$  surface: We have recently demonstrated that co-sensitization of formic acid treated  $TiO_2$  surface not only enhances the dye loading but also reduce the



**Fig. 1.** (a) Scheme representing the various electronic processes at the  $TiO_2$  based DSSC: line 1 shows the dye excitation while line 2 show the electron recombination.  $E_c$  and  $E_f$  represents the conduction band edge (CBE) and Fermi level of  $TiO_2$ ;  $S_0$ ,  $S^+$ ,  $S^*$  represents the ground, excited and oxidized states of the dye, respectively; and  $I^-/I_3^-$  redox potential of electrolyte. (b) Reduction in recombination process via co-adsorbent, which reduces the number of unoccupied  $TiO_2$  site and moves CBE upwards as compared to the untreated  $TiO_2$  surface. (c) Reduction in recombination process through addition of an insulating/semiconducting layer. (d) Reduction in recombination process via co-sensitization of formic acid treated  $TiO_2$  surface.

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