

Interfacial properties of photovoltaic TiO₂/dye/PEDOT–PSS heterojunctions

E.M.J. Johansson^a, A. Sandell^a, H. Siegbahn^a, H. Rensmo^{a,*}, B. Mahrov^{a,b},
G. Boschloo^b, E. Figgemeier^b, A. Hagfeldt^b, S.K.M. Jönsson^c, M. Fahlman^c

^a Department of Physics, University of Uppsala, Box 530, S-75121 Uppsala, Sweden

^b Department of Physical Chemistry, University of Uppsala, Box 579, S-75123 Uppsala, Sweden

^c Department of Science and Technology, Linköping University, S-60174 Norrköping, Sweden

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Abstract

Systems comprising a dense TiO₂ film electrode, a ruthenium polypyridine dye and a PEDOT–PSS (poly(3,4-ethylenedioxythiophene)–poly(4-styrenesulphonate)) film were prepared. The heterojunctions were shown to have photovoltaic properties, with the dye absorbing the light, the TiO₂ acting as an electron conducting material and PEDOT–PSS acting as a hole transport material. A series of dyes was used to investigate their influence on the photocurrent and the photovoltage characteristics of the heterojunction. These results were compared to a photoelectrochemical system in which the PEDOT–PSS was replaced by a liquid electrolyte containing triiodide/iodide redox-couple.

Photoelectron spectroscopy (PES) was used to monitor the interfacial properties of the heterojunction and the investigation points out effects of importance when assembling the materials together to a functional unit. Specifically, it was concluded that the interaction with the dye clearly affects the structure of PEDOT–PSS, both with respect to the surface composition of PSS relative to PEDOT and with respect to the chemical state of the sulphur in the polymers. Moreover, a comparison of the Ru3d and the valence band spectra of the two different interfaces (dye/TiO₂ and dye/PEDOT–PSS) indicates that the energy level structure of the dyes compared to the substrate is different for the two surfaces. Thus, in the combined energy level picture under dark conditions, the energy levels in TiO₂ relative to the energy levels in PEDOT–PSS depend on the dye.

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1. Introduction

Nanostructured dye-sensitized solar cells (DSSC) combined with liquid electrolytes, show promise as low cost alternatives to conventional solid state solar cells [1]. Encapsulation of the liquid electrolyte in these cells leads, however, to some technical difficulties. Replacing the liquid electrolyte in these systems with a solid material may give practical advantages [2]. Conducting polymers could be such a mate-

rial and the combination of dye-sensitized TiO₂ and different types of polythiophenes has been studied by several authors [3–7]. For example, Smestad et al. [5] recently made photoelectrochemical comparisons between non-porous TiO₂ electrodes combined with either polythiophene or with liquid electrolyte. Also, dye-sensitized heterojunctions using PEDOT as a hole-conducting material have been investigated [3,4]. Although the conducting polymers have shown to work as holeconductor material in the DSSC, the total efficiencies of such heterojunctions are yet not as high as the efficiencies obtained for their liquid electrolyte based analogous.

* Corresponding author.

E-mail address: hakan.rensmo@fysik.uu.se (H. Rensmo).

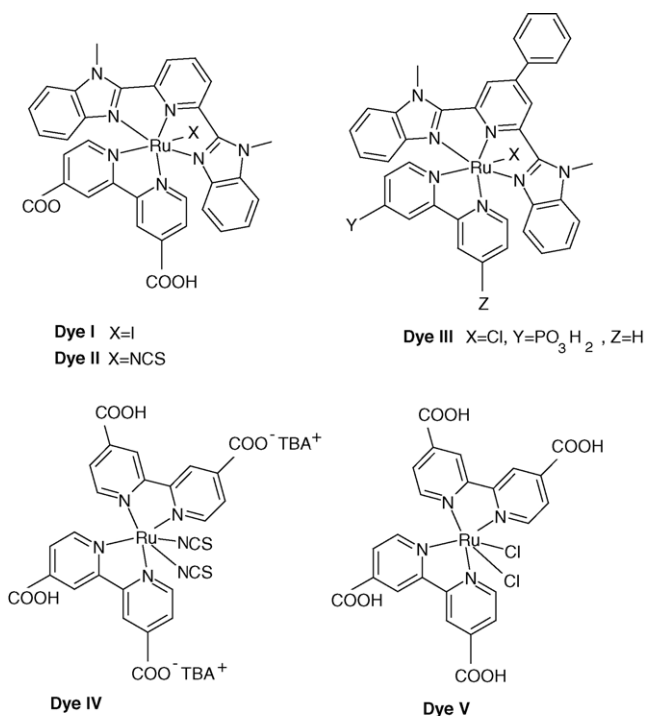


Fig. 1. Molecular structures of investigated dye molecules.

In this study a combination of well-known materials is assembled to form a model system for a solid state DSSC. The system consists of a non-porous (“dense”) TiO₂ electron conducting material, ruthenium–polypyridine dyes as light absorbing materials and PEDOT–PSS (poly(3,4-ethylenedioxythiophene)–poly(4-styrenesulphonate)), as organic hole-conducting material, Figs. 1 and 2. The polymer consists of PEDOT [8,9] which is ‘doped’ (partly oxidized) to become highly conductive and charge compensated with PSS [10]. Since PEDOT–PSS is relatively transparent [11–13] it is an advantage to use it in the DSSC, where the light absorp-

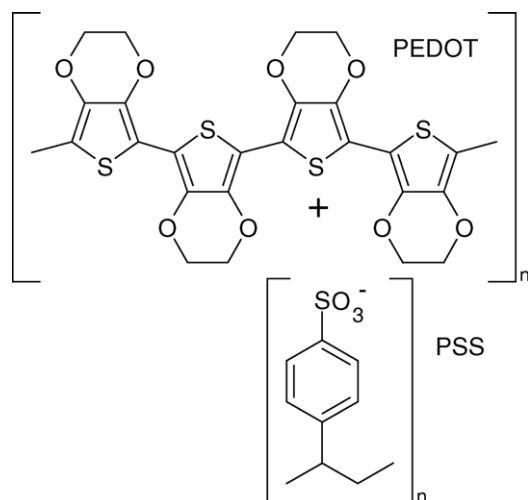


Fig. 2. Molecular structure of PEDOT–PSS.

tion should be carried out mainly by the dye, and the polymer should only be used for transport of electrons to the dye.

The prepared model systems were shown to have photovoltaic properties and the trends were compared to a photoelectrochemical system in which the PEDOT–PSS was replaced by a liquid electrolyte containing triiodide/iodide. The efficiency of the heterojunction largely depends on the electron transfer rates at the interfaces between the different materials (semiconductor, dye and hole-conductor). The transfer rates are closely connected to the energy level matching between the materials at the interface as well as their combined geometrical structure. Insight into the geometrical and the electronic structure of the dye molecules at the different materials is therefore useful in understanding the properties of the heterojunction. Photoelectron spectroscopy (PES) measurements have earlier been used to map the electronic- and molecular structures as well as charge transfer dynamics of dye-sensitized surfaces [14–16]. In order to allow for a PES study of the specific interactions of the interfacing materials in the TiO₂/dye/PEDOT–PSS heterojunction, the dye/TiO₂ and the dye/PEDOT–PSS interfaces were investigated separately. The more detailed investigation of molecular and electronic structures is focused on the dye/PEDOT–PSS interface and the structures are compared to the dye/TiO₂ interface. The investigation specifically points out the effects of importance when assembling the materials together to a functional unit.

2. Experimental

The Ru-dyes used in the experiments are shown in Fig. 1. Dye IV (N719) and V were purchased from Solaronix S.A. [17]. The synthetic procedures of the other dyes are described in Refs. [18–20]. The dye solutions were saturated ethanol solutions (<0.5 mM) except for dye IV (0.5 mM solution in ethanol). All dye-sensitized samples were rinsed by dipping them into ethanol. The thickness of the dye layers were estimated using PES to about one monolayer for all dyes on TiO₂ except for TiO₂ sensitized with dye IV, where the thickness was slightly higher (between one and two monolayers).

The PEDOT–PSS dispersion was provided by Agfa. The following preparation procedure was used for the dye/PEDOT–PSS interfaces: the polymer was spun on FTO glass and then sensitized at room temperature for 5 h in the dye solutions. The samples were then rinsed by dipping them into ethanol. The coverages of the dyes at the substrates were estimated to submonolayers using PES. The sample without dye (a PEDOT–PSS film on FTO glass) was immersed in ethanol for 5 h before the PES measurements. This was done in order to prepare the samples as equally as possible.

For the photocurrent and photovoltage measurements on the TiO₂/dye/PEDOT–PSS system the following preparation procedure was used: the non-porous TiO₂ layer of the model system was made by spray pyrolysis [21] on FTO glass (SnO₂:F on glass). The thickness of the layers was in the order of 300 nm as measured by a Dektak3 surface profile meter.

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