



Thiolate/Disulfide Based Electrolytes for p-type and Tandem Dye-Sensitized Solar Cells



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ABSTRACT

This article presents the first report of a tandem dye-sensitized solar cell employing optically transparent, non-corrosive metal-free thiolate/disulfide based electrolytes and a set of sensitizers with complementary absorption spectra. Sodium 1-phenyl-1H-tetrazole-5-thiolate and its reduced form 5,5'-dithiobis(1-phenyl-1H-tetrazole) (thiolate/disulfide) were used as redox mediators for photocathodic dye-sensitized solar cells (p-DSCs), yielding higher open circuit voltages (285 mV) compared to the more commonly used iodide-based redox couple (226 mV). The herein achieved efficiencies of p-DSCs (0.51 %) and pn-DSCs (1.33 %) employing the thiolate/disulfide-based electrolytes were comparable to p-DSCs and pn-DSCs employing conventional iodide-based electrolytes (0.44 % and 1.19 %).

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Dye-sensitized solar cells (DSCs) with their low projected costs have increasingly attracted attention as an alternative to silicon solar cells [1,2]. The highest reported conversion efficiencies for dye-sensitized photoanodes (n-DSCs) and dye-sensitized photocathodes (p-DSCs) are 13% and 2.5% respectively, under simulated sunlight (1000 W/m², AM1.5 G) [3,4]. These record efficiencies still lag behind the conventional silicon solar cells and some other thin film solar cell technologies. Researchers have been endeavouring to improve the efficiencies of n-DSC and p-DSC by altering the semiconductors, sensitizers and redox couples [3–24]. One attractive approach to improve efficiencies is to construct third generation tandem dye-sensitized solar cells (pn-DSCs). In the n-DSCs, the photocurrent results from dye-sensitized electron injection into n-type semiconductors, e.g. TiO₂, whereas in p-

DSC electron transfer occurs from the valence band of the p-type semiconductor (NiO being the most commonly used to date) to the photoexcited dye [25,26]. These charge separation mechanisms are complementary and provide the opportunity to assemble n-DSCs and p-DSCs in a simple sandwich structure to produce tandem dye-sensitized solar cells (pn-DSCs) [27]. According to Kirchoff's circuit law photovoltages are additive for this type of pn-DSCs, as the photoelectrodes are connected in series. However, the photocurrent is limited by the weaker performing photoelectrode [27,28].

Previously reported pn-DSC efficiencies have been higher than their individual components (n-DSCs and p-DSCs). However, those devices still shared several limitations. The overlapping absorption spectra of the most efficient p-DSC sensitizer (PMI-6T-TPA – see Fig. 1a) with that of the applied n-DSC sensitizers (N719) meant that both photoelectrodes were competing for photons in a similar wavelength range, limiting the attainable pn-DSC photocurrent. Furthermore, the tandem devices were utilizing electrolytes based on the I⁻/I₃⁻ redox couple which absorbs strongly at wavelengths below 500 nm [6].

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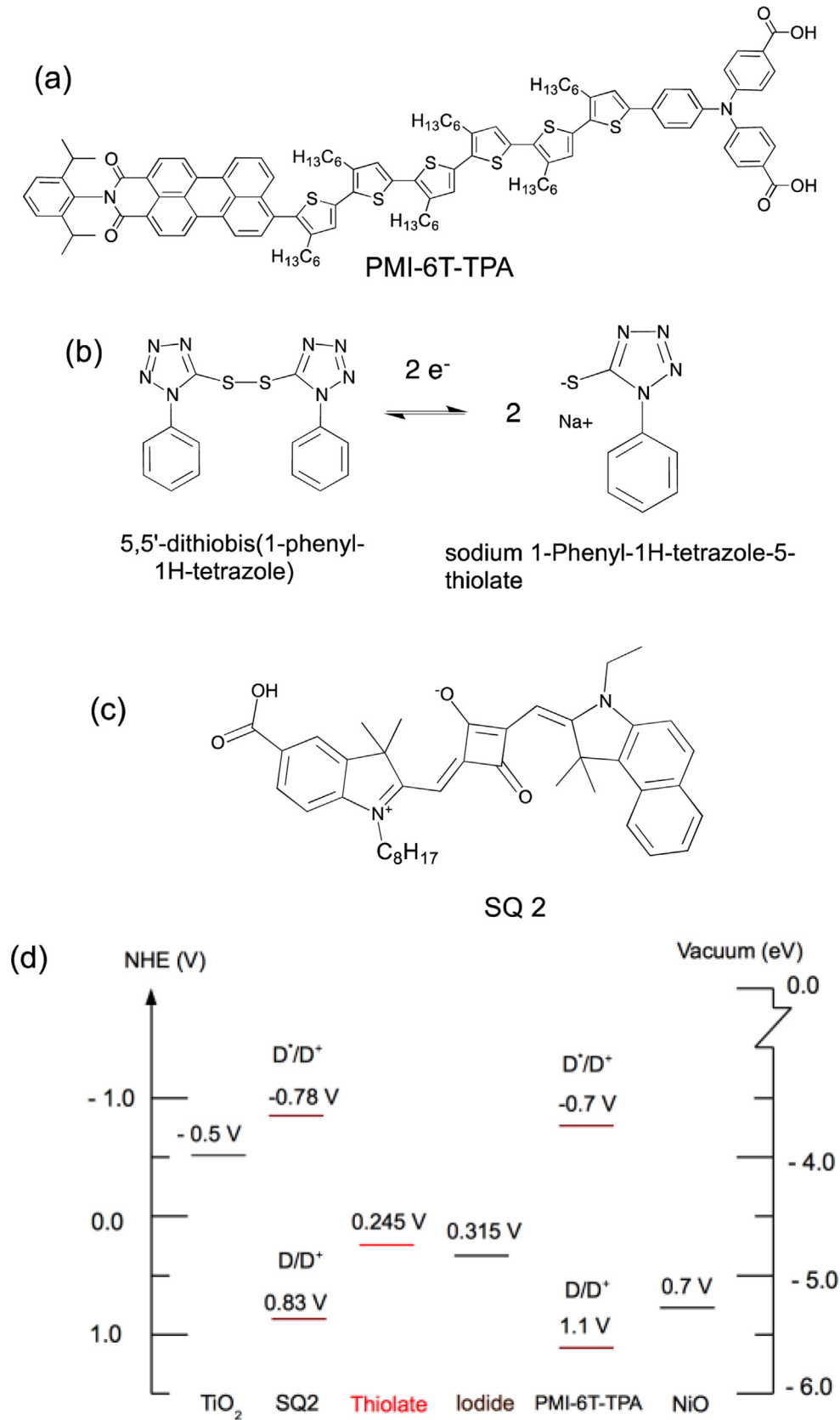


Fig. 1. (a) Structure of the PMI-6T-TPA sensitizer [29]; (b) structures of the oxidized (5,5'-dithiobis(1-phenyl-1H-tetrazole)) and reduced (sodium 1-phenyl-1H-tetrazole-5-thiolate) forms of the redox couple; (c) structure of the SQ2 sensitizer; (d) approximate energy diagram for the components used in the tandem solar cells with the rest potentials of the iodide and thiolate electrolytes (measured according to a literature procedure) [30]. The work functions of NiO (- 5.2 eV) and the sensitizer were measured using photoelectron spectroscopy (PESA) (Figure S1, ESI †) and the conduction band edge of TiO₂ (- 4.0 eV) was taken from the literature [11,29,31].

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