



Solar cells sensitized with near-infrared absorbing dye: Problems with sunlight conversion efficiency revealed in ultrafast laser spectroscopy studies



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ABSTRACT

Complete solar cells sensitized with cyanine dye MK-245, having near-infrared absorption band optimized for tandem devices, are prepared, characterized and studied by time-resolved laser spectroscopy techniques. The photocurrent of the cell correlates with the amount of residual transient absorption signal, pointing to the poor electron injection as the main factor limiting the sunlight conversion efficiency. The measurements show that the lifetime of the excited state is drastically reduced from about 300 ps in solution to single picoseconds when the dyes are attached to titania nanoparticles. It is due to the self-quenching process that effectively competes with electron injection, even under the presence of co-adsorbent. In addition, high concentration of lithium cations in the electrolyte is necessary to improve electron injection quantum yield. The obtained results should be valid for a large class of similar compounds and reveal the main problems in obtaining sufficient performance of near-infrared absorbing dyes.

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1. Introduction

Position of absorption band of the molecules used in dye-sensitized solar cells (DSSC) [1] is one of the most important parameters in the performance and potential application of such photovoltaic devices [2–4]. From among hundreds of different dyes proposed so far for DSSC, the dyes that absorb the red and near-infrared (NIR) part of the sunlight spectrum have been definitely less intensely studied [2,4–6]. It is probably due to lower photovoltage available from the cells sensitized with such dyes, in comparison with those absorbing in shorter wavelengths range. However, the former dyes allow fabrication of semi-transparent devices having unique blue or green colors, important for the decorative and building-integrated photovoltaics (BIPV) application of DSSC. Even more important, the dyes absorbing in the red and NIR range are necessary for tandem devices, the concept that might lead to the improvement of the overall DSSC sunlight conversion efficiency.

The idea of tandem DSSC solar cells takes the benefits of the concept successfully applied in multi-junction crystalline solar cells

[7–9]. The absorbed sunlight spectrum is divided into two (or more) sub-units with active materials of different bandgaps, reducing the limiting thermalization losses taking place in single-bandgap devices. The bandgaps of the absorbing materials needs to be optimized to 1 Sun conditions to fulfill the current matching condition. Recently, we have shown that for the DSSC using iodide based electrolyte, the optimum absorption onset for the dyes in the first and second unit of tandem configuration are 660 nm and 930 nm, respectively [10]. The maximum available efficiency of such tandem configuration is 20%, much more than the current laboratory record for DSSC equal to 13% [11]. However, despite several attempts at different constructions of tandem DSSC [12–20], the obtained efficiencies are still lower than for a single DSSC. The main reason is poor performance of the long-wavelengths part in terms of low incident-photon to current efficiencies (IPCE). IPCE depends on light harvesting efficiency and quantum yields of three partial charge separation processes in DSSC (electron injection, dye recombination and charge collection) [2], and its low values indicate that at least one of these four components of IPCE is significantly less than 100%. Moreover, the absorption bands of dyes used for tandem cells were usually far from optimal with the first absorption onset usually too long, and the second absorption onset too short with respect to the above mentioned ideal ones.

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Therefore, in the present studies we focus our attention on cyanine MK-245 dye (Fig. 1A), whose absorption band between 600 and 950 nm (when attached to TiO₂) matches almost perfectly the tandem cell requirements and which shows a high extinction

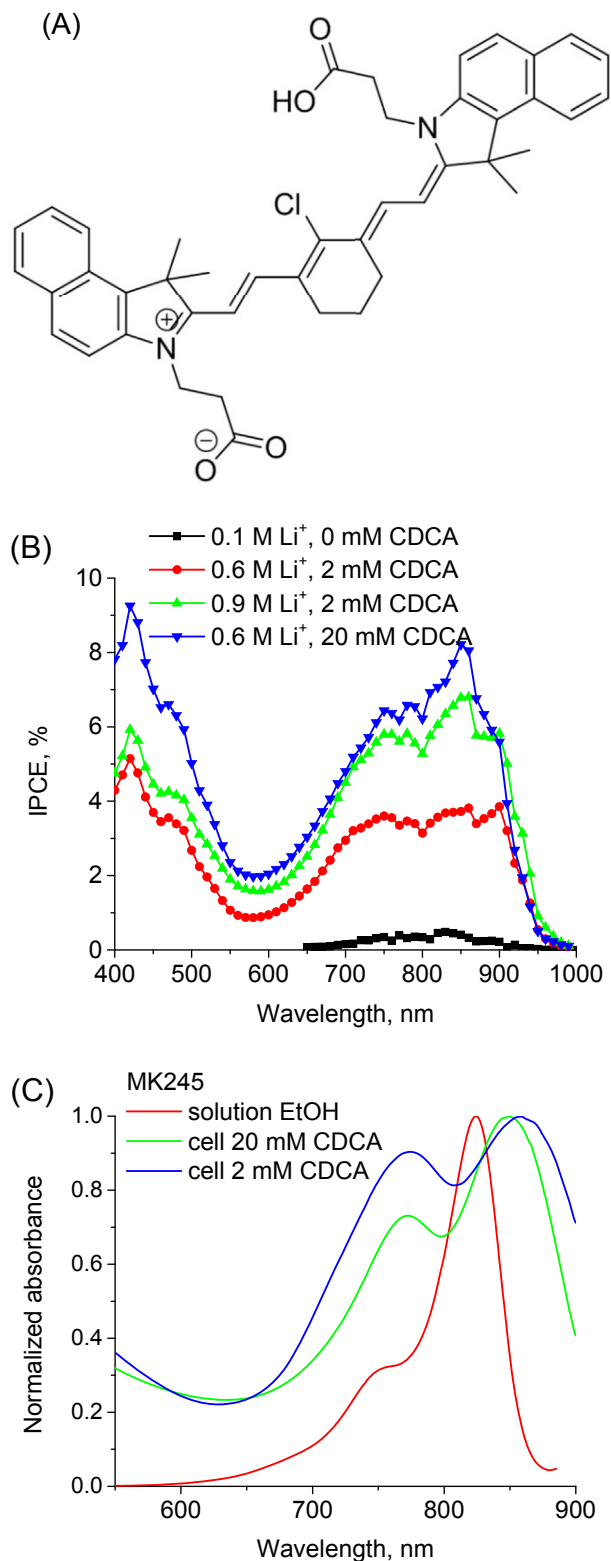


Fig. 1. (A) Chemical structure of MK-245 dye. (B) IPCE spectra of the indicated cells. (C) Steady-state absorption spectra of MK-245 in solution and in solar cells.

coefficient (146 000 M⁻¹ cm⁻¹ in the maximum). To the best of our knowledge, these are the first studies of the cells employing this dye. This dye structure is similar to that of NK-6037 introduced several years ago [21], and differs in the alkyl chain length of COOH and COO⁻ groups. The best efficiencies of the cells with NK-6037 dye (2.3%) were obtained for a very high concentration of co-adsorbent (deoxycholic acid, 0.1 M) used to decrease the dye aggregation, and thus exceptionally thick nanostructured TiO₂ multilayer (25 μm) was required to enable sufficient light harvesting [21]. Electrochemical impedance spectroscopy studies of NK-6037 cells have shown that the collection efficiency (in millisecond range) improves for higher co-adsorbent concentration due to a reduced electron recombination [21].

As will be evidenced below, our cells prepared with MK-245 dye showed tendencies similar to those of the cells with NK-6037 dye. Therefore, we used time resolved spectroscopy applied to ultrafast time scale are important for the cell performance of this group of cyanine dyes. Indeed, we observed that electron injection quantum yield strongly depends on the co-adsorbent concentration and is correlated with the photocurrent of the solar cell. Thus, most probably, for this kind of near-infrared absorbing molecules the charge separation and competing processes occurring in ultrafast time scale are equally or even more important (with respect to slower charge collection) for total sunlight conversion efficiency.

2. Materials and methods

MK-245 dye (3-(2-((E)-2-((E)-3-((Z)-2-(3-(2-carboxyethyl)-1,1-dimethyl-1,3-dihydro-2H-benzo[e]indol-2-ylidene)ethylidene)-2-chlorocyclohex-1-en-1-yl)vinyl)-1,1-dimethyl-1H-benzo[e]indol-3-ium-3-yl)propanoate) was received from Dyenamo AB, Sweden (alternative name is DN-FI07). Titania paste (P25 nanoparticles) was prepared as described earlier [22]. The paste was deposited by doctor-blade technique on fluorine doped tin oxide (FTO) glass plates and heated at 450 °C (TiO₂) for 60 min. The resultant thickness of TiO₂ film was close to 3 μm. Then, sintered electrodes were immersed into a 0.05 M aqueous solution of TiCl₄ stored at 70 °C for 30 min. After that, the electrodes were washed with distilled water and sintered again at 450 °C for 30 min. Then, the films were immersed in a 0.14 mM solution of MK-245 and different concentrations of chenodeoxycholic acid (CDCA): 0 mM, 2 mM and 20 mM) in acetonitrile (ACN): tert-butanol mixture (1:1). The immersion time was about 1 h (at room temperature). The platinumised counter electrode was obtained by spreading a Pt-based solution (Platisol T, Solaronix SA) on FTO glass and annealing it at 450 °C. To make a complete solar cell (the active area of 0.25 cm² was prepared), the counter electrode was assembled with a thermal adhesive film (25 μm Surlyn, Meltronix, Solaronix SA) that acts as separator and sealing element. The cells were filled (by vacuum backfilling method) with one of the following three electrolytes (all in ACN solution): (1) 0.05 M I₂ (99.5%, Fluka), 0.6 M DMPII (1,2-Dimethyl-3-propylimidazolium iodide, 98%, ABCR) and 0.1 M LiI (99%, Aldrich); (2) 0.05 M I₂ and 0.6 M LiI; (3) 0.05 M I₂, 0.6 M LiI and 0.3 M LiClO₄ (99.99% Sigma Aldrich). The electrolytes will be further abbreviated, according to the concentration of lithium cations, as: **0.1 M Li⁺** (electrolyte 1), **0.6 M Li⁺** (electrolyte 2), and **0.9 M Li⁺** (electrolyte 3).

The steady-state UV–visible absorption spectra of MK-245 in solution, MK-245 films and cells were measured on a UV–VIS-550 (Jasco) spectrophotometer (all in transmission mode). A current–voltage characterization of the solar cells and IPCE spectra was performed with a potentiostat (model M101, Autolab) coupled to a photoelectric spectrometer equipped with the option of a solar simulator (Instytut Fotonowy – Photon Institute, Poland). The

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