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Optimization of tandem-structured dye-sensitized solar cell

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ABSTRACT

We optimized tandem-structured dye-sensitized solar cells (T-DSSCs) that consist of a N719-sensitized UV–visible-responsive top cell and a black dye-sensitized near-infrared-responsive bottom cell. The relationship between the thickness (d_T) of TiO_2 film in the top cell and the performance of T-DSSC was examined both for parallel and series T-DSSCs. The optimized parallel T-DSSC gave the photovoltaic conversion efficiency (η) of 10.6% (aperture area; 0.188 cm^2, and total area; 0.25 cm^2) under solar-simulating light conditions (AM 1.5, 100 mW cm^{-2}); this η value was certified as the highest value of the conversion efficiency for T-DSSCs.

1. Introduction

DSSCs sensitized with $[NBu_4]_2$[*cis*-Ru(4,4′-Hdcbpy)$_2$(NCS)$_2$] (**N719**; 4,4′-H_2dcbpy=4,4′-dicarboxy-2,2′-bipyridine) and $[NBu_4]_2$[Ru(Htcterpy)(NCS)$_3$] (referred to as black dye or **BD**; $[NBu_4]^+$= tetrabutylammonium cation; H_3tcterpy=4,4′,4″-tricarboxy-2,2′:6′,2″-terpyridine) have shown high efficiency in the visible and near-IR regions, respectively [1–6]. On the other hands, tandem-structured dye-sensitized solar cells (T-DSSCs; Fig. 1) have been investigated to expand the photo-response into the near-IR region by introducing near-IR-responsive DSSCs as a bottom cell [7–12]. However, the optimal conditions for T-DSSCs have not been investigated to maximize the DSSC's conversion efficiency (η). For the optimal conversion efficiency in such a T-DSSC configuration, the irradiated near-IR light must penetrates into the bottom cell without losing photons at the top cell. Therefore, the optimization of the top cell is an important factor for ensuring the photovoltaic performance of the bottom cell. The effective photon flux for the bottom cell is partly absorbed by the top cell because the absorption spectrum of **N719** partly overlaps with that of **BD**. When the thickness of the TiO_2 film in the top cell (d_T) is enlarged to improve the top cell's conversion efficiency, the efficiency of the bottom cell might decrease. The η of the top cell must be balanced with the bottom cell's performance. First of all, we optimized d_T to improve the efficiency of our T-DSSCs.

We also investigated two kinds of T-DSSCs, i.e., series-connected (ST-DSSC) and parallel-connected T-DSSCs (PT-DSSC). For ST-DSSCs, the short-circuit photocurrent density (J_{sc}) of the top and bottom cells should be same, and for PT-DSSCs, the open-circuit voltage (V_{oc}) of the top and bottom cells should be identical. When these conditions are fulfilled in each case, the conversion efficiency of both ST-DSSC and PT-DSSC could be optimized to give the maximized efficiency. We finally compared η of respective ST-DSSCs and PT-DSSCs for T-DSSCs, obtaining the best conversion efficiency for T-DSSC.

2. Experimental

2.1. Materials

All materials were reagent grade and were used as received unless otherwise noted. **N719** was purchased from Peccell Technologies (Yokohama, Japan). **BD** was purchased from Solaronix S.A. (Lausanne, Switzerland) and was purified by column chromatography using a Sephadex LH-20 column and water as the eluent.

2.2. Dye-Sensitized TiO_2 films

Nanocrystalline TiO_2 films were prepared as described earlier [13,14]. Briefly, nanocrystalline TiO_2 colloids were prepared by hydrolysis of titanium isopropoxide or $TiCl_4$. Then, TiO_2 pastes were prepared by mixing the TiO_2 colloids with α-terpineol and ethyl cellulose. The geometric surface area of each film was

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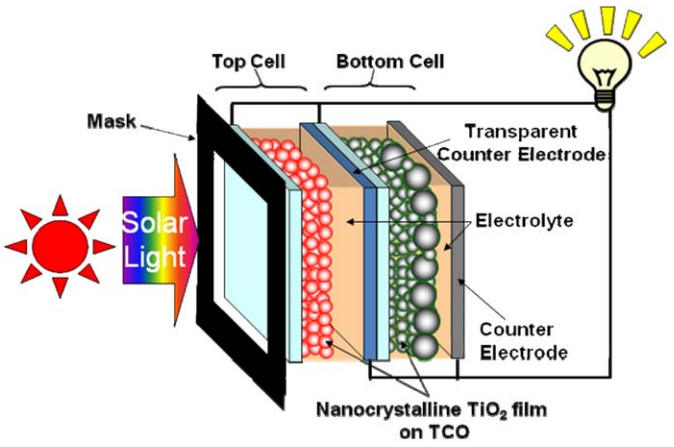

Fig. 1. A schematic structure of the parallel tandem dye-sensitized solar cell (PT-DSSC) used herein.

0.25 cm^2. Film thickness was controlled by means of screen-printing and was measured with an Alpha Step 500 profiler (KLA Tencor Co., California, USA). The films of the top cells were composed of a transparent layer. The particle size was ca. 25 nm and a narrow size distributions as same as the Ref. [14]. The films of the bottom cells were composed of a transparent layer and a scattering layer (6 and 17 μm thickness, respectively) in order to effectively absorb the light in transparent layer by the light back-scattering of the scattering layer [13]. The particle size of the transparent layer was ca. 20 nm, whereas the scattering layer was composed of small and large particles with diameters of 20 and 300 nm, respectively. After an annealing of the transparent TiO_2 films of the top cells at 500 °C, the films of the top cells were submerged in a solution of 3×10^{-4} M **N719** in a mixture of acetonitrile and *tert*-butyl alcohol (volume ratio, 1:1) at room temperature for 48 h. The films of the bottom cells were also annealed at 500 °C, and were immersed in ethanolic **BD** solution (2×10^{-4} M) containing 2×10^{-2} M deoxycholic acid at room temperature for 20 h. We determined the extent of **BD** or **N719** adsorption by measuring the amount of the dyes' respective ruthenium complexes that had adsorbed on the films. To obtain these measurements, the ruthenium complexes were desorbed from the TiO_2 films into a 0.01 M NaOH aqueous solution, and the UV–visible absorption spectrum of this solution was measured with a UV–visible spectrometer (UV3101PC, Shimadzu Co., Kyoto, Japan).

2.3. Methods

The single DSSC consists of a dye-coated TiO_2 film electrode, an electrolyte solution and a Pt counter electrode [13,14]. For the top cells, a transparent Pt counter electrode was prepared by the calcination of a solution of H_2PtCl_6 spin-coated on transparent conductive glass (TCO, Japan Sheet Glass 10 Ω cm^{-2}) which consists of F-doped SnO_2 on glass. Pt was sputtered on TCO to create Pt counter electrodes for the bottom cells. All the cells then were sealed with a thermally adhesive film (HIMILAN, Du Pont-Mitsui Polychemicals Co., Tokyo, Japan). The UV–visible transmittance of the top cells was measured with the UV–visible spectrometer. For photovoltaic measurements, we used a two-light-source solar simulator with Xe and halogen light sources (WXS-80C-3, 100 mW cm^{-2}, Yamashita Denso Co., Tokyo, Japan) to simulate the AM 1.5 spectrum. A black metal mask with an aperture area of 0.175 cm^2 was placed over the cells during irradiation. An anti-reflection glass coated by thin layer of Ta_2O_5, SiO_2 and MgF_2 was placed on the front glass cover of the cells. We adjusted the irradiation power to 100 mW cm^{-2} by using secondary reference solar cells, which consisted of single crystal Si cells equipped with Schott KG5 (3 mm thick) or KG3 (1 mm thick) filters that were calibrated by the Japan Electrical Safety and Environment Technology Laboratories—Research Division.

3. Results and discussion

3.1. Photovoltaic properties of the top cells

We investigate short circuit photocurrent density (J_{sc}^{TOP}), open circuit voltage (V_{oc}^{TOP}) and fill factor (FF^{TOP}) of **N719**-sensitized solar cells with increase of thickness (d_T) of TiO_2 films to optimize T-DSSCs consisting of **N719**-sensitized top cells. The J_{sc}^{TOP} value increased with increasing the d_T as well as an increase of conversion efficiency (η) as shown in Fig. 2(a). The solid line in Fig. 2(a) shows a correlation between J_{sc}^{TOP} and d_T that is calculated from LHE (see Eqs. (1)–(3) in Section 3.3). These facts are explained as due to increase of light harvesting efficiency (LHE)

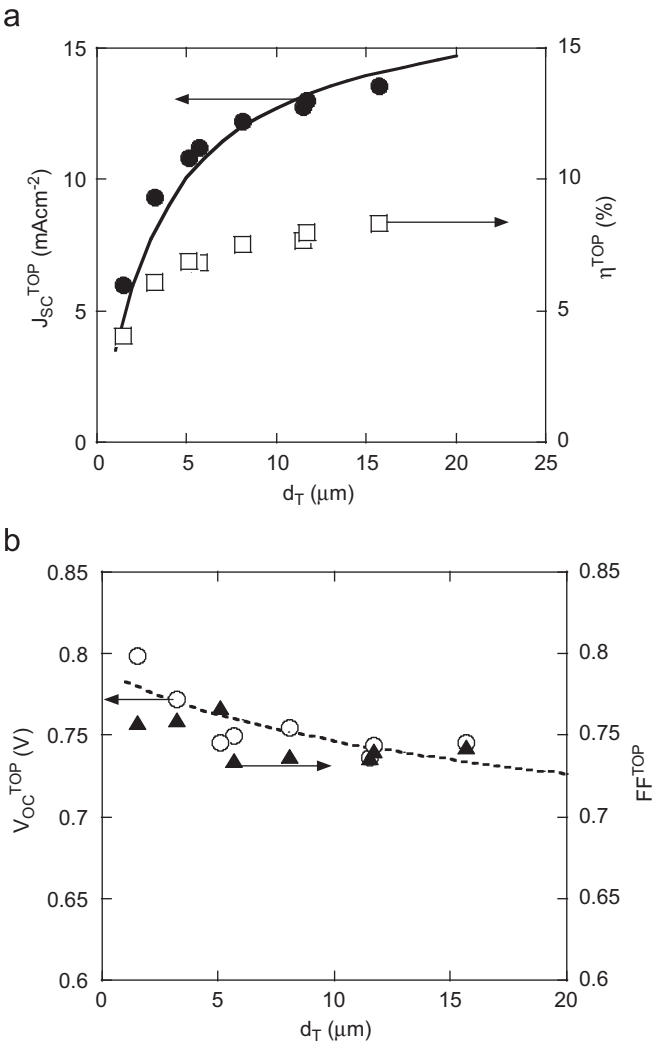

Fig. 2. (a) J_{sc}^{TOP} (●) and η^{TOP} (□) of the top cells with varying d_T. The solid line was J_{sc}^{TOP} calculated from Eq. (1), Eq. (2), and Eq. (3). (b) V_{oc} (○) and *FF* (▲) of the top cells with varying d_T. The dotted line is the best-fitting line of the relationship between V_{oc}^{TOP} and d_T, as shown in Eqs. (5) and (6). The arrows in figure shows *y* axis of each plot.