# **Development of Integrated Tandem Dye Sensitized Solar Cells**

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# Abstract

In this work we demonstrate a novel Integrated Tandem Dye Sensitized Solar Cell in which the working electrode of the top cell is similar to that of a single absorber DSSC and consists of  $TiO_2$ -NPs sintered on TCO/Glass substrate, while the bottom cell is TCO-free and constructed in such a way that the working electrode and the common counter electrode co-exist on the same substrate.

## 1. Introduction

Dye-sensitized solar cell (DSSC) [1] is a third generation photovoltaic device that offers an attractive efficiency to cost ratio. However the improvement of the energy conversion efficiency up to 15% is highly desirable for the industrial application of the DSSC. Unfortunately, the intrinsic light absorption characteristics of the organic dyes are different from those of inorganic semiconductors. As a result, the attempts to develop a single dye absorber that can cover the broad range of the solar spectrum are highly challenging.

An alternative approach to broad the spectral response of the device is to use a tandem absorber where two mesoporous working electrodes are sensitized with different dyes, which are sensitive to different parts of the solar spectrum. At present, two possible ways to construct a tandem DSSC were reported. First is an "n-n" mechanical stack of two independently fabricated DSSCs with a semi-transparent TCO/Pt counter electrode for the top cell [2-4]. Obviously, such a tandem device keeps all electrical losses typical for the single absorber DSSC and introduces significant optical losses, because light has to pass through three TCO layers (one of which is semitransparent TCO/Pt) before it arrives to the bottom cell. Although the mechanical stack tandem DSSC is easy to fabricate, the optical losses significantly diminishes the benefits of having the tandem absorber. The second possible structure is an "n-p" tandem DSSC based on a p-type transparent mesoporous counter electrode that is sensitized with a special "p-type" dye [5,6]. Although this concept is promising, the currently known p-type transparent oxides have very poor conductivity. As a result, the up-to-date efficiency of the p-type sensitized DSSCs is extremely low.

In this work we demonstrate a novel integrated tandem dye sensitized solar cell (IT-DSSC) which realizes a third possible way to construct the DSSC tandem. The IT-DSSC is a two-substrate sandwich device. The working electrode for the top cell is similar to the single absorber DSSC while the bottom cell is TCO-free and constructed in such a way that the working and the counter electrodes co-exist on the same substrate (see Fig. 1).



Fig. 1. Schematic illustration of the Integrated Tandem DSSC. SEM images of the nanoporous alumina (NPA) membrane (a) and Ti/Al/Ti porous contact layer (b).

## 2. Experimental

The fabrication of the bottom cell started from the deposition of an adhesion layer on glass substrate by reactive r.f. magnetron sputtering from pure Ti target (99.99%) in a mixture of Ar:O<sub>2</sub> (1:1) at a base pressure of 1.1 Pa. After the initial deposition of a 5 nm thin  $TiO_2$  layer the oxygen flow was gradually reduced to zero to enrich the surface of the adhesion layer with Ti. Then the W and Al thin films with the thicknesses of 500 nm and 1500 nm were deposited consequently in pure Ar at the base pressure of 0.7Pa from W (99.95%) and Al (99.99%) targets, respectively. The Al film was converted into nanoporous alumina (NPA) membrane by anodization in 0.15M phosphoric acid at 10 °C at a constant potential of 120 V in a two-electrode configuration with the Al layer, as the anode, and a platinum mesh, as the counter electrode. The anodization process resulted in the formation of nanoporous alumina (NPA) membrane with the through-hole structure while the surface of W was oxidized locally under the

nanopores. The dissolution of the tungsten oxide was conducted in acetate pH=7 buffer solution and was immediately followed by the electroless deposition of Pt nanorods inside the NPA by immersion of the sample into a 0.05M solution of  $PtCl_2$  in acetate pH=7 buffer.

The fabrication of the working electrode of the bottom cell started from sputtering of a Ti/Al/Ti film on the top of the NPA membrane with the overall thickness of 250 nm. Then the mesoporous TiO<sub>2</sub> was fabricated on the Ti/Al/Ti film by screen printing and sintering of a paste with TiO<sub>2</sub> nanoparticles at 450 °C. The working electrode of the top cell was fabricated on the TCO/glass substrate by the same screen printing and sintering procedure. The top and the bottom mesoporous electrodes were of the same thickness of 10  $\mu$ m. The sensitization was carried out in 2x10<sup>-4</sup>M solution of N719 in acetonitrile and  $2x10^{-4}$  solution of the BD in ethanol for the top cell and the bottom cell, respectively, during 48 hours at room temperature. The IPCE and I-V characteristics of the individual cells and the tandem were measured in an open cell configuration with iodine electrolyte. The TiO2 paste, the dyes and the iodine electrolyte were purchased from Solaronix.



Fig. 2. SEM images of the top cell (a) and the bottom cell (b) of the integrated tandem DSSC device.

#### 3. Results and Discussion

The bottom cell resembles the concept of the TCO-less DSSC with a back contact developed by Han [7]. However in contrast to Han we managed to integrate the working and the counter electrodes on one substrate by using the NPA membrane (Fig. 2b). We can clearly see the Pt nanorods grown on the W underlayer inside the NPA membrane and Ti/Al/Ti film with sintered TiO<sub>2</sub> mesoporous layer. The Ti/Al/Ti film replicates the morphology of the NPA membrane (Fig. 1b) and has nanoholes through which the iodine electrolyte can penetrate to the Pt nanorods underneath. The IPCE spectra and I-V characteristics measured for the top cell, the bottom cell and the tandem are shown on the Figures 3a and 3b, respectively. The

detailed characterization of the individual cells and the tandem device will be presented during the conference. We would like to emphasize that in this configuration of the tandem device the top and the bottom cells are connected in parallel and thus result in a combined Jsc (15.3 mA/cm<sup>2</sup>) and averaged Voc (0.72 V). The overall conversion efficiency of the tandem is 8.4%.

## 3. Conclusions

We demonstrated a novel integrated tandem DSSC that consist of a N719 dye sensitized top cell and a black dye sensitized bottom cell with the overall conversion efficiency of 8.4%. The integrated tandem DSSC is a two-substrate sandwich device with conventional working electrode for the top cell and TCO-free working electrode for the bottom cell that is integrated with the common counter electrode on one substrate.



Fig. 3. (a) IPCE and (b) I-V characteristics of the top cell (green), the bottom cell (red) and the tandem device (black).

#### Acknowledgements

We acknowledge financial support from the Ivan Pulyui Scholarship Program of Ukraine, Core Research for Evolutional Science and Technology (CREST) of the Japan Science and Technology Agency (JST) and the New Energy Development Organization (NEDO) of Japan.

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