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## Scaling of the flexible dye sensitized solar cell module

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#### ABSTRACT

Dye sensitized solar cell (DSSC) has attracted tremendous attention over last two decades and is currently regarded as a promising alternative to the conventional flexible photovoltaic (PV) market. Thin, light-weight and flexible DSSC module enables deployment on curved surfaces which significantly widens the application space. The critical challenge in advancing flexible DSSC technology remains in the up-scaling of module dimensions. Herein, we report advancement achieved in developing large-scale flexible DSSC modules of the dimensions 100 mm × 100 mm with conversion efficiency of 3.27%. A facile binder-free TiO<sub>2</sub> paste was synthesized to fabricate firm TiO<sub>2</sub> photoanode on plastic substrate at low temperature. Cold isostatic press (CIP) technique was utilized to not only improve the quality of film but also to increase the thickness of TiO<sub>2</sub> film. Based on binder-free TiO<sub>2</sub> paste, flexible DSSC module with dimension of 100 mm × 100 mm was fabricated and demonstrated for mobile phone charging application under indoor light condition.

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#### 1. Introduction

Dye sensitized solar cell (DSSC) is regarded as the third generation of solar cell and has attracted much attention owing to low fabrication cost and environment-friendly synthesis process [1–3]. Conversion efficiency is the most important indicator for solar cell to evaluate its capability for converting light into electricity. Since the report from O'Regan and Grätzel in 1991, the conversion efficiency of DSSC has been increased from 7.1% to 13% [4,5], exceeding the average efficiency of amorphous silicon solar cell. A typical DSSC uses fluorine doped tin oxide (FTO) glass as the transparent conductive substrate. Majority of studies in literature on high efficiency DSSC have been conducted on glass substrate [6-8]. The employment of FTO glass could enable high temperature process and ensure superior durability. However, the drawbacks of glass based DSSC are also apparent, such as increased weight and dimensions, and reduced strength which limits the application of DSSC modules. It is obvious that light-weight and flexible DSSC will have more opportunities in practical scenarios. Flexible DSSC fabricated on conductive plastic substrate or metal foil renders solar cell with conformabilities to uneven surfaces. It can be uniquely designed for specific platforms to satisfy the special shape requirements, such as attachment to the surface of windows, clothing and portable electronic devices. Furthermore,

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http://dx.doi.org/10.1016/j.solmat.2016.07.021 0927-0248/© 2016 Elsevier B.V. All rights reserved. the fabrication of flexible DSSC is suitable for roll-to-roll process, enabling the implementation of mass production and further reduction of manufacturing cost [9–11].

In order to fabricate flexible DSSC, several techniques have been employed to enable low temperature synthesis of layers. Yamaguchi et al. utilized a compression method to prepare TiO<sub>2</sub> film on flexible polymer substrate. Owing to the light confinement effect of large size particles in TiO<sub>2</sub> film, the conversion efficiency of 7.4% was achieved [12]. Chiu et al. employed 2-step electrophoretic deposition (EPD) technique to prepare TiO<sub>2</sub> film on polymer substrate. Due to the scattering layer, the device showed a conversion efficiency of 6.63% [13]. Recently, Liang et al. reported a new architecture of flexible DSSC. They utilized flexible plastic capillary tube as an outer cover, and Pt microwire counter electrode and TiO<sub>2</sub> nanotube photoanode were assembled in the plastic tube to form the flexible solar cell. An impressive conversion efficiency of 9.1% was derived from this tubular flexible solar cell [14]. Despite high efficiency of flexible DSSC obtained in these prior studies, the research efforts were confined to small size cell which limits the application domain. Current challenge in DSSC technology is related to the up-scaling of module dimensions in order to provide ability to meet the desired power requirements. Generally, the preparation of DSSC module involves synthesis steps that will not be factors in fabrication of small size cell, and thus it is questionable whether the high efficiency would be maintained as the DSSC module is developed with a large number of cells connected in series or parallel architecture. There have been some attempts in literature on addressing the challenges

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related to DSSC module [15-19], however, the performance remains below the application requirement [20]. In addition, most of the prior research on DSSC module has focused on glass module and only limited number of investigations have been conducted on flexible DSSC module such as conductive plastic substrate based module. In this paper we provide significant advancement in realization of DSSC flexible module by systematically addressing all the material and processing parameters that limit the integration of material layers on low temperature substrates. A facile binder-free TiO<sub>2</sub> paste was developed for low temperature synthesis of high quality TiO<sub>2</sub> photoanode. In order to improve the performance of flexible DSSC module, the factors limiting the conversion efficiency of low temperature synthesized materials were studied. Based on the performance improvement of the binder-free paste, flexible DSSC modules with dimension of 100 mm  $\times$  100 mm were fabricated, characterized and optimized.

#### 2. Experimental section

#### 2.1. Materials

FTO glasses (14  $\Omega$ /sq) were provided by Nippon Sheet Glass Co. Ltd. Commercial high temperature TiO<sub>2</sub> paste (Ti-Nanoxide T/SP), large titania particle paste (Ti-Nanoxide R/SP), low temperature TiO<sub>2</sub> formulation (Ti-Nanoxide T-L), hot melt sealing film (Meltonix 1170-60) and dye sensitizer (N719) were purchased from Solaronix. Flexible conductive substrate of indium tin oxide/polyethylene naphthalate (ITO/PEN, <15  $\Omega$ /sq) was purchased from Peccell. Liquid electrolyte (IoLiLyte SB-163) was purchased from IoLiTec. TiO<sub>2</sub> nanoparticles (Degussa, P25), 2-propanol, acetonitrile, tert-butanol, NaBH<sub>4</sub>, H<sub>2</sub>PtCl<sub>6</sub> and conductive silver paste were purchased from Sigma-Aldrich.

#### 2.2. Binder-free $TiO_2$ paste preparation

P25 paste was first prepared by dissolving P25 nanoparticles in the mixed solvent of tert-butanol and DI water (v:v=2:1), followed by stirring for 2 h and ultrasonic dispersing for 30 min to obtain a homogenous paste. The binder-free TiO<sub>2</sub> paste for the flexible DSSC was prepared by blending the P25 paste with the commercial T-L TiO<sub>2</sub> slurry (w:w=10:1), which was followed by stirring for 2 h, ultrasonic dispersing for 30 min, and ball milling for 48 h.

#### 2.3. DSSC module preparation

#### 2.3.1. Glass DSSC module

The silver fingers were first printed on FTO glass by using screen printer (HMI, MSP-485). After drying the printed silver fingers, the TiO<sub>2</sub> photoanode was prepared by screen printing of T/SP paste and then the light scattering layer with R/SP paste was printed. The printing was followed by annealing at 450 °C for 1 h. Pt counter electrode was prepared by spreading 10 mM solution of H<sub>2</sub>PtCl<sub>6</sub> in terpineol on FTO glass followed by annealing at 500 °C for 1 h. Next, the silver fingers were printed on the Pt coated electrode. The assembly of DSSC module was done by laminating photoanode and counter electrolyte was injected through the tiny holes in the counter electrolyte followed by sealing the holes with epoxy resin.

#### 2.3.2. Flexible DSSC module

The silver fingers were first formed on ITO/PEN substrate by screen printing and then drying at 120 °C for 1 h. Next, the  $TiO_2$  film was prepared on ITO/PEN substrate with the binder-free  $TiO_2$ 

paste by doctor blade method and drying at room temperature for 30 min. After that the TiO<sub>2</sub> coated ITO/PEN was sealed in scotch thermal laminating pouches and compressed using CIP process (Avure Tech Inc.) at pressures of 15 kPsi for 3 min. The dye sensitization was performed by immersing flexible TiO<sub>2</sub> film in 0.5 mM solution of N719 dye in acetonitrile and tert-butanol (v:v=1:1) at room temperature for 12 h. The flexible counter electrode was prepared by chemical reduction method as follows: 10 mM H<sub>2</sub>PtCl<sub>6</sub> in 2-propanol solution was uniformly spread on the surface of ITO/PEN followed by annealing at 120 °C for 1 h. After that, the H<sub>2</sub>PtCl<sub>6</sub> coated ITO/PEN electrodes was immersed in 10 mM NaBH<sub>4</sub> aqueous solution at room temperature to reduce the Pt cations to metallic Pt nanoparticles on the surface of ITO/PEN. Silver fingers were printed on the Pt deposited ITO/PEN, and the counter electrode was dried at 120 °C for 1 h. The assembly of flexible DSSC module was done by laminating photoanode and counter electrode with the hot melt sealing film at 110 °C. Lastly, the liquid electrolyte was injected through the tiny holes in the counter electrolyte, followed by sealing the holes with epoxy resin.

#### 2.4. Characterization

X-ray diffraction (XRD) on TiO<sub>2</sub> film was conducted by Philips Xpert Pro x-ray diffractometer (Almelo, The Netherlands). UV–vis absorption spectra were taken using UV–vis spectrophotometer (U-4100, Hitachi). The morphology and thickness of TiO<sub>2</sub> films were examined by scanning electron microscopy (SEM, Quanta 600 FEG, FEI). A solar simulator (150 W Sol 2A<sup>TM</sup>, Oriel) was employed to provide air mass (AM 1.5) illumination of 100 mW cm<sup>-2</sup>. J-V characteristics of DSSC were measured in dark condition and under illumination by using Keithley digital source meter (Model 2400).

#### 3. Results and discussion

#### 3.1. Development of binder-free TiO<sub>2</sub> paste

The plastic ITO/PEN substrate cannot endure the temperature higher than 150 °C. This leads to the exclusion of post-annealing process for the preparation of flexible TiO<sub>2</sub> photoanode. Thus, the TiO<sub>2</sub> paste for flexible DSSC cannot use binder materials that need high temperature process. Low temperature sintering presents challenge in the synthesis of TiO<sub>2</sub> thin film with high quality surface and high adhesion with substrate. In this study, we developed a facile binder-free sticky TiO<sub>2</sub> paste by combining P25 paste and T-L paste, denoted as PT paste. P25 paste recipe comprises of dissolution of P25 nanoparticles in a mixed solvent of water and tert-butanol. This mixture can reduce the surface tension of paste to improve the  $TiO_2$  film adhesion on the flexible substrate [21,22]. The P25 paste exhibited high viscosity as demonstrated in Fig. 1 (a) where the paste remains at the bottom of the vial without flowing. However, upon addition of T-L paste, the viscosity decreases dramatically and the paste readily flows in the vial. The viscosity of TiO<sub>2</sub> paste is related to the surface interaction force. The high viscosity of P25 paste is due to the attractive forces dominant between the nanoparticles resulting in large agglomerates. The steric stabilizing repulsive force could suppress the nanoparticle agglomerates and facilitate the dispersion of nanoparticles resulting in low viscosity of paste [23]. With the addition of acidic T-L TiO<sub>2</sub> slurry into the P25 paste, the surface of the TiO<sub>2</sub> nanoparticles will be positively charged according to Eq. (1) [24,25]. The surface charging suppresses the agglomeration and promotes dispersion of TiO<sub>2</sub> nanoparticles in paste.

 $Ti-OH+H^+ \rightarrow Ti-OH_2^+$ 

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