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Flexible dye-sensitized solar cell fabricated on plastic substrate by laser-detachment and press method

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ABSTRACT

This report shows that flexible dye-sensitized solar cell can be fabricated by a laser-detachment and press method where the TiO_2 electrode typically sintered on glass source substrate is detached by a laser pulse and then is firmly adhered to the conductive plastic substrate by applying a high pressure. The cells fabricated by this process exhibited 36–43% smaller photocurrent and efficiency than the conventional glass cells with directly coated TiO_2 electrodes. It was attributed to the lowered dye coverage and electron diffusion length, both of which originate from the press-induced reduction of TiO_2 porosity. A maximum efficiency of 5.68% was obtained for the plastic cell. Bending of the electrode led to 20% loss of the current density and efficiency. However, no further performance degradation was observed even when the bending cycle was increased to 100, 300, and 500 times. This indicates that the bending-induced degradation of TiO_2 electrode on the plastic substrate takes place at the first bending.

1. Introduction

Dye-sensitized solar cells (DSSCs) have been under intense investigation in many institutes as potential low-cost photovoltaic devices [1–5]. In conventional DSSCs, the TiO_2 photo electrodes are prepared on conductive glass substrates using printable paste, followed by annealing at 450–500 °C [1]. The sintering process is essential to remove the organic additives contained in paste and achieve better electrical connection between the nanoparticles. Nowadays, the efficiency of DSSCs reaches 12%, almost catching up with that of amorphous Si solar cells [6]. Replacing the glass substrates with flexible ones not only reduces the weight and cost of the devices but also extends the application areas. In particular, DSSCs fabricated on the bendable and light-weight plastic substrates are suitable as small-scale power generation units in a modular fashion, thus receiving much attention [7–10]. However, the thermal instability of plastic substrates precludes high-temperature sintering processes and the use of organic binders. This is the main reason why plastic DSSCs have lower efficiencies than conventional glass-based cells. A lot of efforts have been made to solve this problem. Hagfeldt et al. [11] have introduced a press method for the low-temperature preparation of nanostructured TiO_2 electrodes. Using porous TiO_2 films prepared by this method, they achieved an efficiency of 5.5% under 10 mW/cm^2 (0.1 sun). Miyasaka et al. [12] reported on the low-temperature preparation of mesoporous TiO_2 film using a binder-free paste consisting of nanocrystalline TiO_2 and interparticle binding agent. A maximum efficiency of 5.8% was obtained under 100 mW/cm^2 (1 sun). Yamaguchi et al. [13] achieved more efficient plastic DSSCs by the press method. The conversion efficiency could be improved by adjusting the press condition and thickness of TiO_2 layer, along with the surface treatment of plastic substrate. A validated efficiency of 7.6% was reported.

Many of the methods reported to date rely on the low-temperature preparation of electrodes [7,9–15]. In DSSC, the inter-particle necking is inevitable for efficient charge transport. Any low-temperature processing is likely to cause incomplete necking between TiO_2 nanoparticles, which limits the maximization of device performance. Durr et al. [9] suggested a lift-off technique. In this process, the TiO_2 film is first coated onto a thin Au layer deposited on glass substrate. After sintering, this TiO_2 layer is removed from the substrate by dissolving the Au layer and is then transferred into a conductive plastic substrate by application of high pressure. They achieved an efficiency of 5.8% under 100 mW/cm^2. Considering that the electrode was sintered by standard high-temperature process, the obtained efficiency is not so high. They found that the major loss when compared with the typical glass system is due to a reduction in the short-circuit current density (J_{sc}). Even though it was attributed to the lower visible transmittance of indium tin oxide (ITO)-covered polyethylene terephthalate (PET), some other factors may be responsible for the reduced current density. This article presents a laser transfer technique in which the deposition and dissolution of Au layer is unnecessary. In order to investigate which step determines the

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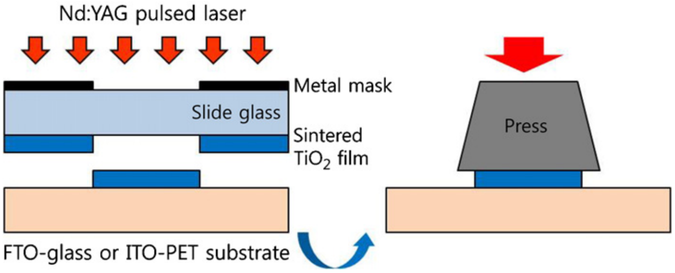

Fig. 1. Schematic of the electrode transfer process.

reduction of current density and efficiency (η), the characteristics of conventional DSSCs fabricated on glass substrate, cells transferred onto glass, and those transferred on plastic substrate are comparatively analyzed, along with the stability of plastic DSSCs against bending.

2. Experimental procedure

The TiO_2 electrodes were first prepared on glass source substrates using a commercial paste (Ti-Nanoxide D/SP, Solaronix). They were coated by the doctor blade method using 3 M scotch tape as the spacer. The coated films were dried at 125 °C for 30 min and then sintered at 500 °C for 30 min in ambient air, finally being $TiCl_4$ treated. The $TiCl_4$ treatment was done by immersing the TiO_2-coated substrate in a 40 mM $TiCl_4$ aqueous solution. After being kept for 30 min at 70 °C in the $TiCl_4$ solution, the sample was rinsed with distilled water and then sintered again for 30 min at 500 °C. Fig. 1 shows a schematic of the electrode transfer process. The TiO_2 electrode prepared on the source substrate (bare slide glass) was made in contact with the receiver substrate ultimately used as the transparent front electrode of DSSC. Fluorine-doped tin oxide (FTO)-coated glass (TEC 8, 10 Ω/sq., 2.2 mm thickness) and ITO-covered PET (EMI-15 Ω/sq., 175 μm thickness) were used as the conductive receiver substrates. The TiO_2 electrode was detached from the source substrate by exposing it to a laser beam incident onto the TiO_2 film from the backside of the source substrate. An ultraviolet Nd:YAG pulsed laser ($\lambda = 355$ nm, pulse width < 5 ns, repetition rate = 10 Hz, maximum average power = 1.8 W, output beam diameter = 0.9 cm) was employed as the laser source and a single pulse was incident in the detachment process. This method is fundamentally based on the laser-induced thermal desorption [16–21] and is different from the conventional laser ablation technology. Nanoparticle or nanostructured film adhered to the substrate can be desorbed when the thermo-elastic force, caused by a rapid thermal expansion of the film resulting from pulsed laser irradiation, exceeds the adhesive force (predominantly van der Waals force) between the film and the substrate. In this process, a bare slide glass was used as the source substrate so that the incident laser beam is mostly absorbed by TiO_2. When the pulse energy was too high, the TiO_2 film was fragmented. With careful control of the pulse energy, however, the film could be detached without fragmentation.

Since the as-detached film exhibited poor adhesion to the receiver substrate, it was adhered to the receiver substrate by application of high pressure using a hydraulic press (Specac, GS15011, UK). A pressure of 212 kg/cm^2 was applied for 15 s at room temperature. The electrode size for cell fabrication is 4 mm × 4 mm. After pressing, the electrodes were immersed into an ethanol solution that contained 0.5 mM of D719 dye (Everlight, Taiwan) and were held for 24 h. The Pt counter electrode was coated by dropping a diluted H_2PtCl_6 solution into the FTO-covered glass, followed by curing for 15 min at 400 °C. For the plastic counter electrode, a Pt layer was deposited onto ITO-PET substrate by DC sputtering (Cressington sputter coater-108 auto). Two substrates coated with the electrodes were put together and sealed using a sealant. The liquid electrolyte (Iodolyte AN-50) was injected through the hole punched in the counter electrode. This hole was immediately sealed by soldering a sealant for the glass cell and putting a sticky tape for the plastic cell. The cell efficiency was measured using a solar simulator (PEC-L01, Peccell Technology, Japan) on the condition of AM 1.5, 100 mW/cm^2 (150 W Xenon light source, class A spectral match, irradiation uniformity $\leq \pm 3\%$) and a Si photodiode was used for the reference calibration. The electron lifetime and diffusion coefficient were measured using intensity-modulated photovoltage spectroscopy (IMVS) and intensity-modulated photocurrent spectroscopy (IMPS), respectively (Ivium modulight module, $\lambda = 635$ nm, diode-to-sample distance = 1.5 cm, power = 52 mW, Netherlands). The structure and morphology of TiO_2 film was investigated by field-emission scanning electron microscopy (JEOL JSM-7001 F, 30 kV).

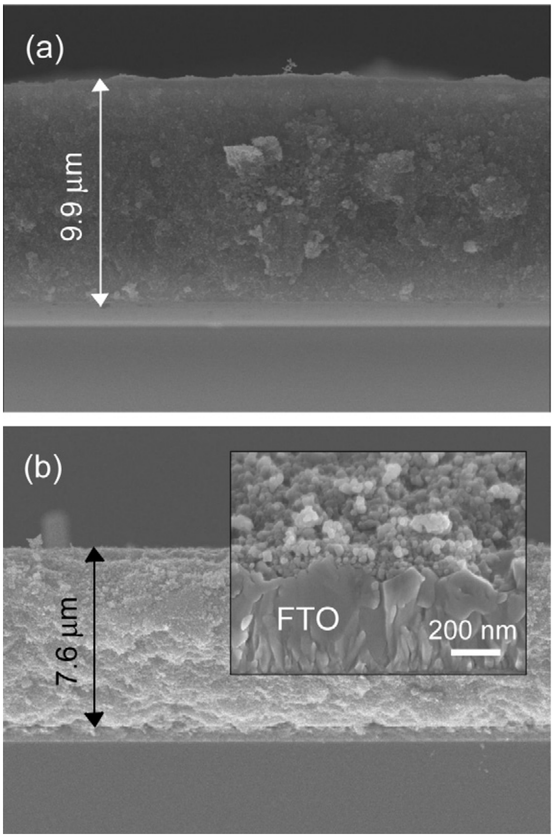

Fig. 2. (a) Cross-sectional image of a TiO_2 layer sintered on glass source substrate. (b) TiO_2 layer taken after being transferred onto FTO-glass substrate. The inset shows the interface between TiO_2 and FTO.

3. Results and discussion

Fig. 2(a) shows the cross-sectional image of a TiO_2 layer sintered on source substrate. The thickness was about 10 μm. After the detached layer was pressed onto FTO-glass substrate, the thickness was reduced, as shown in Fig. 2(b). The interface image shows that the transferred electrode is firmly adhered to the receiver substrate (inset of Fig. 2(b)). As the layer thickness decreased, the internal morphology of electrode was somewhat modified as well. Fig. 3(a) and (b) compare the internal morphology of TiO_2 electrode before and after transfer. The average particle size increased probably due to the inter-particle agglomeration. In addition, individual particles became more tightly packed with the disappearance of sporadically observed big pores. This indicates that the transferred electrode has