

Nanostructured TiO₂ films for dye-sensitized solar cells

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Abstract

A new strategy was attempted to fabricate photoelectrode of dye-sensitized solar cells through a reassembly technique (exfoliation and restacking) to increase solar activity. A random hybridization between exfoliated layered titanate and TiO₂ (anatase) nanoparticles resulted in a ‘house-of-cards’ structure, which might increase the mesoporosity and surface area of TiO₂ film. In the XRD patterns of the present nanocomposite, no (00 *l*) peaks could be seen due to the random hybridization between layered titanate and TiO₂ nanosol particles. According to the N₂ adsorption–desorption isotherms, the resulting nanohybrids are fairly porous with a high specific surface area ($S_{\text{BET}} = \sim 190 \text{ m}^2/\text{g}$, mean pore size = 6 nm), which leads to an efficient dye adsorption. A solar energy to electricity conversion efficiency (η) of the present nanocomposite film is about 100 times higher than that of TiO₂ nanoparticle one under the standard AM 1.0 irradiation condition, suggesting that the mesoporous nature of TiO₂ film would play an important role for efficient photovoltaic cell performance.

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1. Introduction

Interest in dye-sensitized nanocrystalline TiO₂ based solar cells (DSSCs) has grown considerably in recent years for fundamental and applied perspectives. Although there are many other semiconducting metal oxides such as SnO₂, ZnO, Nb₂O₅, CeO₂, and SrTiO₃, TiO₂ has been the most widely used one for solar cell. It exhibits efficient light harvesting capacity as well as low production cost. Particularly, intense research has been undertaken on the textural properties of nanocrystalline TiO₂ films since they play a key role in light harvesting of solar cell [1–3]. TiO₂ is sensitized for visible light by the dye chemisorbed on the surface of TiO₂ film as a monolayer. In fact, a current hot issue is the fabrication of mesoporous TiO₂ films with high surface area to accommodate more sufficient dye absorption as well as to facilitate transport of electron and electrolyte in DSSC.

The films for the photoelectrode with high surface area have been fabricated exclusively with nanoparticles of anatase TiO₂.

In the conventional DSSC, to avoid formation of micropores, a sponge-like mesoporous structure is prepared from anatase nanocolloids hydrothermally in the presence of polymer as a binder [4]. However, the thermal treatment to remove the organic binder significantly diminished the surface area because of aggregation of nanoparticles. Although this kind of nanocrystalline TiO₂ films exhibits relatively much higher surface area than traditional nonporous one, its textural properties such as surface area and pore structure remain still to be enhanced. Recently, an approach to mesoporous anatase network was attempted with the ordered mesoporous TiO₂ synthesized by surfactant-template method [5,6]. Unfortunately, they suffer from low thermal stability and poor crystallinity in spite of high surface area and regular mesoporosity.

We here attempted to fabricate ‘house-of-cards (HOC)’ structured TiO₂ nanohybrid in order to overcome the disadvantage of both nanoparticle-derived and ordered mesoporous TiO₂ films. This structure offers the salient features for dye-sensitized nanocrystalline TiO₂ solar cell. HOC nanohybrid consists of anatase nanoparticles attached over the exfoliated layered titanate nanosheets in random arrangement. Unlike simple pillaring of semiconductor nanoparticles into the interlayer spaces of layered titanate, which led to highly microporous structure, HOC nanohybrid could develop high

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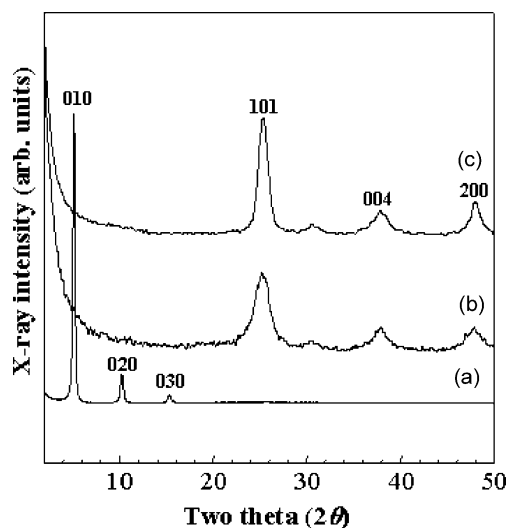


Fig. 1. Powder X-ray patterns for (a) TBA-intercalated layered titanate, (b) as-prepared HOC nano hybrid, and (c) HOC nano hybrid with the heat-treatment at 450 °C for 1 h.

mesoporosity without any significant decrease in high surface area of anatase nanoparticles. The surface area and mesoporosity could be optimized for idealized chemisorption of dye molecules. The resulting hybrid material was systematically investigated by X-ray diffraction (XRD), scanning electron microscopy (SEM), along with a preliminary test for the performance of DSSC.

2. Experimental section

Cesium titanate, $\text{Cs}_{0.67}\text{Ti}_{1.83}\text{O}_{4.17}$, was prepared by heating a stoichiometric mixture of Cs_2CO_3 and TiO_2 at 800 °C for 20 h. The corresponding protonic form, $\text{H}_{0.67}\text{Ti}_{1.83}\text{O}_{4.17} \cdot \text{H}_2\text{O}$, was

obtained by reacting cesium titanate powder with 1 M HCl aqueous solution at room temperature for 3 days. The layered protonic titanate was exfoliated by intercalation of TBA (tetrabutylamine) molecules, as reported previously [7]. On the other hand, TiO_2 nanocolloid solutions were prepared by hydrolysis of titanium isopropoxide in presence of HNO_3 aqueous solution (375 ml) and peptizing at 80 °C for 8 h [8]. The HOC structured nanohybrid was prepared by slowly mixing the exfoliated titanate with the TiO_2 nanoparticle at room temperature followed by aging for 24 h with continuous stirring. The resulting viscous slurry was deposited onto $\text{SnO}_2:\text{F}$ conducting glasses substrate (Asahi glass Co., ~ 10 ohm/sq, 80% transmittance in the visible) using the simple doctor blade technique, and annealed at 450 °C for 1 h in air. The annealed film was immersed in absolute ethanol containing 3×10^{-4} M $\text{Ru}[\text{LL}'(\text{NCS})_2]$ ($\text{L} = 2,2'$ -bipyridyl-4,4'-dicarboxylic acid, $\text{L}' = 2,2'$ -bipyridyl-4,4'-ditetrabutylammoniumcarboxylate; Solaronix) for 24 h at room temperature. The dye-coated electrode were rinsed with absolute ethanol and dried under N_2 stream. Transparent Pt counter electrodes with small holes were prepared by spreading a drop of 5 mM hexachloroplatinic acid in 2-propanol over a conducting glass, followed by heating at 450 °C for 30 min in air. The Pt electrode was placed over the dye-coated electrode, and the cell edges were sealed with 30 μm -thick Surlyn (Dupont, grade 1702). The redox electrolyte consisted of 0.8 M 1,2-dimethyl-3-hexyl-imidazolium iodide (C6DMI) and 40 mM I_2 in acetonitrile was introduced into the cell by capillary action. The active area of the cell was about ~ 0.36 cm^2 .

The crystal structures for the samples were studied by powder X-ray diffraction (XRD) measurements using Ni-filtered $\text{Cu K}\alpha$ radiation ($\lambda = 1.54056$ Å) with a Philips PW 1830. The electrode microstructure was examined using by a scanning electron microscope (SEM, Hitachi S-4300).

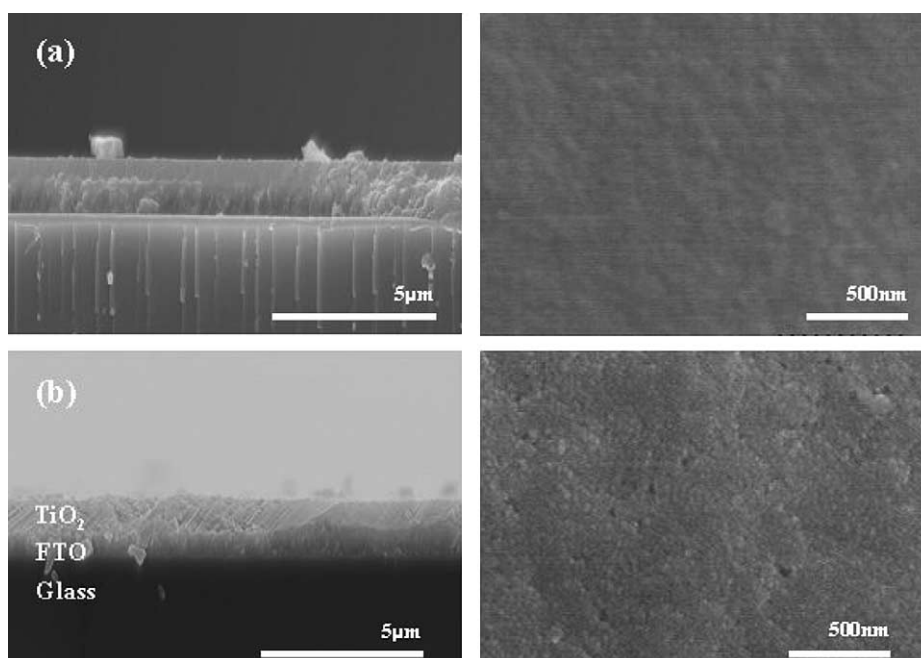


Fig. 2. Scanning electron microscopy (SEM) images for (a) TiO_2 nanoparticles and (b) the HOC nano hybrid: the cross-sectional view (left) and the top view (right).

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