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Effect of photoelectrode morphology of single-crystalline anatase nanorods on the performance of dye-sensitized solar cells

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ABSTRACT

TiO_2 terpineol-based pastes with nanorods (NRs) of over 25 μm thickness have been prepared for the photoactive electrodes of the dye-sensitized solar cells (DSSCs). The NRs, with a length of approximately 80 nm and an aspect ratio of about 3, are made by a two-step hydrothermal process. They have the single crystalline anatase structure and can be dispersed well in water and ethanol. With a high thermal stability and larger surface area (47.2 $m^2 g^{-1}$) than commercial TiO_2 particles (P25, 39.1 $m^2 g^{-1}$), the well-dispersed anatase NR films with aggregate-free morphology are transparent. For the photocurrent–voltage measurements, the NR cell exhibits high short-circuit photocurrent (J_{SC}) under 1 Sun AM 1.5 simulated sunlight due to the higher surface area and transmittance. The open-circuit voltage (V_{OC}) of NR films is not obviously reduced with incremental thickness, which results from the one-dimensional single crystalline structure of NR due to less surface defects. As compared with the P25 cell, DSSCs made with NRs have a higher fill factor (FF) because of the uniform void spaces. An enhancement of conversion efficiency from 4.88% for P25 to 5.67% for NR is achieved. The P25 particles are incorporated in NR films as light-scattering centers, while the R1P1 containing 50 wt.% of P25 has a high V_{OC} and FF as compared with P25, but the J_{SC} is still lower than that of the NR.

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1. Introduction

The dye-sensitized solar cell (DSSC) is one of the most promising low-cost, high-efficiency, and environmentally friendly solar cells [1]. The DSSC is composed of a high absorption efficiency sensitized-dye, high-surface-area TiO_2 film on a conducting glass substrate, a counter electrode with platinum coating, and an I^-/I_3^- redox electrolyte solution. The dynamics compete with DSSC for the initial events of electron injection and dye regeneration leading to photo-induced charge separation, which occurs in the femtosecond to nanosecond time scale; however, the electron transport in the TiO_2 film and interfacial recombination on the TiO_2 surface due to I_3^- are in milliseconds or even seconds [2]. Therefore electron transport and recombination are the key factors affecting the efficiency.

In order to increase electron transport and decrease interfacial recombination, one-dimensional (1-D) nanostructured materials (nanotubes, nanorods, and nanowires) are prepared. It is anticipated that they will have a single crystalline structure to facilitate incremental electron transport in the photoelectrode and less surface defects for reducing recombination on the TiO_2 surface. However, the nanowire (NW) TiO_2 prepared by the Shuzi group need to be treated with Nb in order to increase the thermal stability [3], while the nanorods prepared by the Motonari group need block-copolymers to be added to maintain the shape because of its poor thermal stability [4].

Other than the thermal stability issue, the dispersion problem of the one-dimension TiO_2 particles needs to be surmounted. Single-crystalline anatase NW TiO_2 prepared by the Yiying group is blended with a small amount of nanowires to increase the conversion efficiency, but the high ratio of the NWs blending displays a disappointing conversion efficiency as a consequence of poor dispersion [5]. The photoelectrode films made by well-dispersed TiO_2 increase the transmittance and the surface area for incremental current density, and compact the film structure to reduce the pore size [6,7].

One-dimension nanorod (NR) TiO_2 can be expected not only to increase electron transport but also reduce the surface defects for reductive recombination. In this paper, one-dimension NR TiO_2 with a single crystalline anatase structure is prepared by hydrothermal process and have excellent thermal stability even when the sintering temperature is higher than 500 °C. Furthermore, this NR TiO_2 does not coagulate or agglomerate with each other and can be easily dispersed, resulting in a large surface area for adsorbing more dye to promote high short-circuit photocurrent (J_{SC}). The anatase NR films made by well-dispersed NR are transparent and possess more compact structure, which results in more contact and uniform void spaces for ion diffusion in the TiO_2 photoelectrode. An enhancement of light-to-electric conversion efficiency for NR (5.67%) is achieved as compared with commercially-available TiO_2 powder (P25, Degussa) (4.88%).

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In addition, P25 particles are incorporated in NR films as a light-scattering center to increase the optical length. The resultant effects on J_{SC}, open-circuit voltage (V_{OC}) and fill factor (FF) are studied.

2. Experimental section

2.1. Preparation of TiO_2 particles and paste

The established standard procedures for the preparation of TiO_2 nanorods are conducted according to the method reported by Tadao Sugimoto [8]. First, titanium isopropoxide (TIPO) and triethanolamine (TEOA) are mixed at a molar ratio of TIPO:TEOA = 1:2 to form the compound Ti^{4+}, which is stable against hydrolysis at room temperature. This is followed by the addition of distilled water and 1 M NaOH to make the final solution of 0.5 mol dm^{-3} in Ti^{4+} and a pH = 11. The final solution is placed in a Teflon-lined autoclave and aged at 100 °C for 24 h for gelation; then, the temperature is increased to 150 °C for 72 h to nucleate and grow TiO_2 nanorods.

The TiO_2 paste is prepared by employing Graltzel's method [9]. First, the NR TiO_2 is dispersed in ethanol with ultrasonic bath and then mixes with terpineol and ethyl cellulose as binders. After removing the ethanol with a rotary-evaporator, the final NR paste is prepared. However, the P25, via $TiCl_4$-fumed gas synthesis, is a powder and is difficult to disperse in ethanol. We add the shaker process to help in the dispersion of P25 before being put in ultrasonic bath and the same mixes with terpineol and ethyl cellulose. The P25 paste is made by removing the ethanol. The R1P1 paste is made by mixing 50 wt.% NR and 50 wt.% P25. The other processes are the same with the above-mentioned.

2.2. Preparation of TiO_2 electrode

The fluorine-doped SnO_2 conducting glasses are cleaned by surfactant, acetone and isopropanol, and a spin coating Ti precursor solution in order to make good mechanical contact between the TiO_2 layer and conducting glass. Then, the TiO_2 film is made by doctor blade and heated to 450 °C 30 min. The layer thickness is determined by Alpha-step IQ surface profilometer (Tencor Instruments, USA).

2.3. Nanorod and electrode characterization

Characterization of the nanorods is made by X-ray diffraction (XRD) at 40 kV and 30 mA (Rigakue, RINT2000), Raman Spectrometer, transmission electron microscopy (TEM) at 80 kV (JEOL, JEM-1200EX), high resolution-transmission electron microscopy (HR-TEM) (FEI Tecnai G2), isotherm of nitrogen adsorption at 77 K (Micromerit ASAP 2020), and Dynamic light scattering (DLS) (Nano ZS, Malvern, UK) to get TiO_2 crystalline structures, particle size, lattice spacing, surface area and particle size distribution respectively. Characterization of the electrodes is made by field emission scanning electron microscopy (FE-SEM) at 10 kV (JEOL, JSM-6700F) and ultraviolet–visible light Spectrophotometer (Beckman, DU-800) to get TiO_2 film morphology and light transparency, respectively.

2.4. Fabrication of dye-sensitized solar cells and photoelectrochemical measurements

After sintering at 450 °C and cooling to 80 °C, the TiO_2 electrodes are dye-coated by immersing them into dye solutions at 60 °C for 12 h, which consisted of 0.3 mM N719 dye (Solaronix) in EtOH (99.9%). The

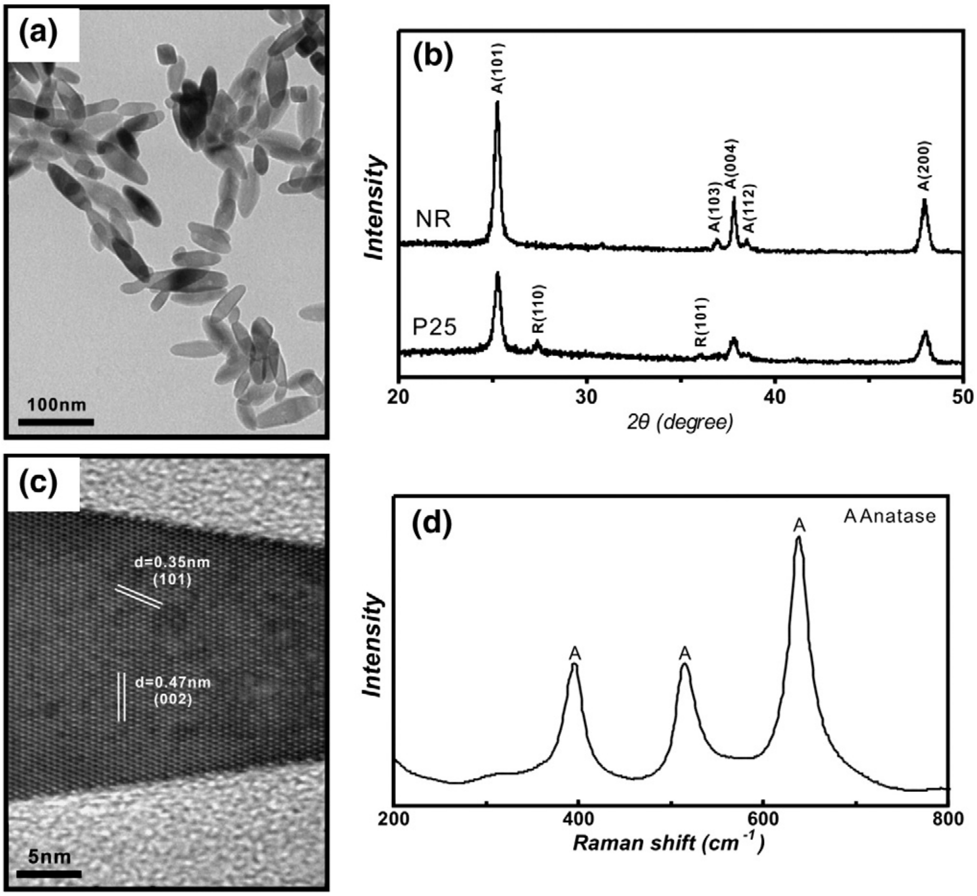

Fig. 1. (a) TEM images, (b) XRD pattern, (c) High-resolution TEM image and (d) Raman spectrum of the NR.

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