APPLICATION OF SINGLE CRYSTALLINE ANATASE TITANIA NANOTUBES FOR DYE-SENSITIZED SOLAR CELLS Supachai Ngamsinlapasathian,

Yusuke Murata, Susumu Yoshikawa, and Motonari Adachi Institute of Advanced Energy, Kyoto University Uji, Kyoto 611-0011, Japan

Highly efficient dye-sensitized solar cells become feasible using single crystalline anatase titania nanotubes (TiNTs) for semiconductor thin film instead of titania nanoparticles.

We used laurylamine hydrochloricde (LAHC) / tetraisopropylorthotitanate (TIPT) modified with acetylacetone (ACA) system. TIPT-ACA solution was mixed with 0.1 M LAHC aqueous solution at 40 $^{\circ}$ C, and the solution was stirred until the solution became transparent. Then, temperature was changed to 80 $^{\circ}$ C. After 3 h, the solution became gel. After 5 h, the hard gel became weak. After 25 to 72 h, the solution became a white gel with a yellow liquid thin layer on the gel.

Figure 1 shows a TEM image of TiNTs in a gel. We can see long nanotubes (diameter 10 nm, length 200 nm) as well as short ones (diameter 10 nm, length 30 nm). Electron diffraction patterns of these nanotubes show the Debye-Scherrer rings of anatase. A TEM image of the calcined sample shown in Figure 2 shows lattice image of the nanotubes, which clearly show that each nanotube is single crystal of anatase. The observed spacings between lattice images agreed with anatase 101 spacing 0.35 nm. These findings confirmed that the titania nanotubes were single crystalline anatase.

Preparation of titania thin films was carried out as follows. We used gel samples before calcinations. Surfactant molecules in TiNTs can be removed by washing with alcohol. We controlled the alcohol content by the centrifugation speed. The titania sample was applied to the conducting glass (Asahi glass) and distributed with a glass rod. After drying, the sample was calcined at 450° C for 30 min. Dye was introduced to the titania thin films by soaking the film for 20 h in a 3×10^{-4} M solution of the ruthenium dye in ethanol. N3 produced by Grätzel's group was used as the dye. We used 0.03 M iodine and 0.3 M lithium iodide in NMO/acetonitrile the electrolyte. The as photocurrent-voltage characteristics were measured using a potentiostat by irradiating with simulated solar light, i.e., AM 1.5 100 mW/cm². The cell size was 1 cm² or 0.25 cm². Titania particle P-25 was used as reference titania for comparison.

Figure 3 shows the short-circuit current density obtained from the cell made of TiNTs, together with those of P-25, against the film thickness. In the thin film region, the photocurrent density of the cell made of TiNTs was 2 times to 3 times higher than that of P-25. This is remarkable point of TiNT cell. The high photocurrent density is attributed to the high electron transfer property in the titania film made of TiNTs. Figure 4 shows an example of photocurrent-voltage characteristics of a cell with double layer titania film, i.e., thick upper film made of TiNTs including 5 % P-25 and thin lower film made of only TiNTs. The obtained short-circuit photocurrent density was 18.4 mA/cm², open-circuit voltage was 0.63 V, fillfactor was 0.62, and light to electricity conversion was 7.2 %. These data promise realization of highly efficient dye-sensitized solar cells using TiNTs as semiconductor thin film through

further improvement.



Anatase crystal

Figure 1. TEM image of titania nanotubes in gel sample.



Bar: 17.4 nm





Figure 3. Relationship between the photocurrent density and the film thickness.



Figure 4. An example of photocurrent- voltage characteristics of a cell made of TiNT double layer.