Formation of Bilayered TiO_2 Films on FTO Glasses for Dye-sensitized Solar Cells: An In-situ XRD Study

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Dye-sensitized solar cell (DSSC) based on TiO_2 film developed has been extensively regarded as an efficient, cost-effective and promising alternative to silicon-based photovoltaic devices. A nanocrystalline TiO_2 particle layer is deposited onto a transparent conducting oxide (TCO) glass substrate as the anode of the DSSC. In this system, TiO_2 film should provide sufficient surface roughness for dye adsorption, serve as a expressway to facilitate the electron transport, and effectively confine the incident light to enhance the light-harvesting. The conventional method to prepare TiO_2/TCO electrode requires sophisticated procedures for layer-by-layer TiO_2-coating. Herein, we report a facile process to fabricate TiO_2/TCO electrode by direct deposition of the TiO_2, which is obtained from hydrolysis of $TiCl_4$, onto the TCO. The influence of $H_2SO_{4(aq)}$ concentration on the phase of titania formed in the $TiCl_4/HNO_3$ hydrothermal system was studied using BL01C2 and BL17A1 at NSRRC.

Figures 1(A)-(E) show the in-situ XRD patterns of 0.6 M $TiCl_{4(aq)}$ hydrolyzed in 5 M $HNO_{3(aq)}$ at 100°C with $H_2SO_{4(aq)}$, in the range of 0 to 80 mM. As shown in Figs. 1, the formation of anatase (A) and/or rutile (R) phases of titania can be tunable by adjusting $H_2SO_{4(aq)}$ concentration. Without addition of $H_2SO_{4(aq)}$, it results a pure rutile phase. As increasing in $H_2SO_{4(aq)}$ concentration, it tends to form anatase. With the $H_2SO_{4(aq)}$ concentration above 40 mM.

We then prepared the TiO_2-based photoanode for DSSCs by direct growing TiO_2 film onto FTO glass via $TiCl_4/HNO_3$ hydrothermal system under various H_2SO_4 concentration. The photovoltaic performances of DSSC were carried out under an Oriel AM 1.5 solar simulator with an intensity of 100 mW/cm^2. Figure 2 shows the obtained photocurrent density-voltage characteristics. The maximum photoconversion efficiency of 4.11% was obtained for the one which was prepared in 60 mM $H_2SO_{4(aq)}$, and the titania film appeared to be a bilayered microstructure with a rutile TiO_2 uplayer on the top of anatase TiO_2 underlayer (as shown in the inset of Fig. 2). We have demonstrated that a tunable layered structural titania film can be directly fabricated by $TiCl_4/HNO_3$ hydrothermal system for efficient DSSC application.

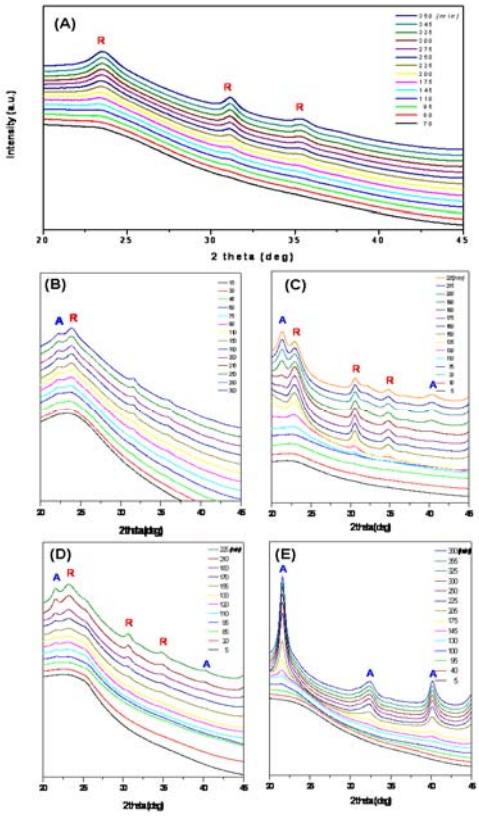

Fig. 1: In-situ synchrotron XRD studies of $TiCl_4/HNO_3$ aqueous system, where [$TiCl_4$] = 0.6 M, [HNO_3] = 5 M with various [H_2SO_4] : (A) 0 mM, (B) 20 mM, (C) 40 mM, (D) 60 mM, and (E) 80 mM at 100°C. (A and R denote anatase and rutile, respectively).

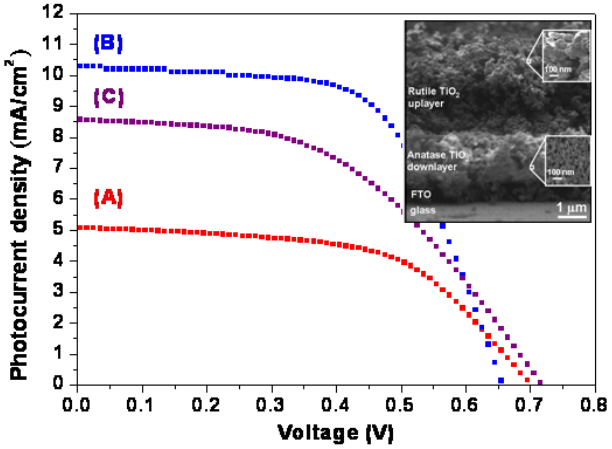

Fig. 2: Photocurrent density-voltage charactiristics of DSSCs based on the photoanodes formed in $TiCl_4/HNO_3$ aqueous system, where [$TiCl_4$] = 0.6 M, [HNO_3] = 5 M with various [H_2SO_4] : (A) 0 mM, (B) 60 mM, and (C) 80 mM at 100°C. (The inset is FESEM image of sample (B)).