Influence of Electrolyte Concentration on Photovoltaic Performance of DSSCs Based on TiO_2 Nanotube Array

Sheng-Qiang TONG[a,*], Jie-Jie CHENG[b]

School of Science, Wuhan University of Technology, Wuhan, China

[a]837859422@qq.com, [b]405095788@qq.com

*Corresponding author

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Abstract. The present work reports a double-layer composite film made of TiO_2 nanotube arrays (TNT-arrays) as the overlayer and TiO_2 nanoparticle (P25) as the underlayer as the photoanode of dye-sensitized solar cells (DSSCs). The TNT-arrays were fabricated by two-step anodization process. In order to further improve the conversion efficiency of DSSCs, TNT-arrays were optimized by changing the anodization conditions. The photoelectric conversion properties of DSSCs with different electrolyte concentration were investigated and compared, and the highest conversion efficiency of 4.20% was achieved in double-layer photoanode at the electrolyte concentration of 0.3wt%.

Introduction

As a new type of thin film solar cell, dye-sensitized solar cells (DSSCs) have attracted widespread concern in the world due to its high photoelectric conversion efficiency, low production cost, and simple manufacture. [1-3]Generally, DSSCs comprises three components: nanocrystalline TiO_2 film photoanode, redox electrolyte, and counter electrode. [4, 5]As one of the most important components in DSSCs, the nanocrystalline TiO_2 film covered by a monolayer of dye molecules has significant influence on photoelectric performance of DSSCs. [6]Recently, due to the good and uniform microstructure, TiO_2 nanotube arrays (TNT-arrays) are widely applied in the field of energy storage, photocatalysis, and photovoltaics.

Herein, we report a novel double-layer TiO_2 photoanode consisting of TiO_2 nanotube arrays and TiO_2 nanoparticle (P25) on a FTO glass substrate. The TNT-arrays were fabricated by two-step anodization process. The influences of anodization parameters on the nanotube array structure and morphology were studied in the NH_4F/ethylene glycol electrolyte via one-step anodization. As a result, DSSCs based TNT/P25 double-layer photoanode achieved the highest conversion efficiency of 4.20% when the electrolyte concentration is 0.3wt%.

Experiment

TiO_2 nanotube array films were obtained by anodization method. The first anodization was carried out in electrolyte solution containing different NH_4F (0.1wt%, 0.3wt%, 0.5wt%, 1.0wt%) while voltage 50V was applied for 19h. Following the first anodic oxidation, TiO_2 nanotube arrays were gained on the surface of pure Ti plate. Moreover, the processes of second anodization were explored on the Ti substrates for 1 h under the same conditions as the first anodization. Finally, the amorphous TiO_2 nanotube arrays films were obtained.

In order to prepare the double-layer films, TiO_2 nanoparticle viscous paste was coated onto the FTO glass substrate via doctor-blade technique, and the as-prepared TNT-arrays were placed onto the P25 underlayer immediately. After being annealed for 30 min at 500 ^oC in air, the electrodes were synthesized through dissolving as-prepared photoelectrodes into a 0.5mM ethanol solution of N719.

In addition, the electrolyte solution was injected into interspace between the photoanode and the counter electrode, and the assembled cells were tested immediately.

Results and Discussion

The surface morphology of TiO_2 nanotube arrays and the cross-sectional SEM of double-layer composite film were shown in Fig.1. Fig.1 (a) and (b) shows that TNT-arrays film is apparently formed by highly ordered nanotube which is opened on the top but closed on the bottom. Fig.1 (c) shows the cross-sectional SEM image of double-layer film with TNT-arrays overlayer (ca.15um in thickness) and P25 underlayer (ca.9um in thickness), and these two layers are in good attachment. As the underlayer of photoelectrode, P25 will enlarge contact area between the TiO_2 film and FTO glass. TiO_2 nanotube arrays overlayer provides good channels for electron transfer which greatly improves the photoelectric properties of DSSCs. However, there will be some covering on the surface of nanotubes when high concentration electrolyte solution was applied to anodic oxidation process (see Fig.1 (d)). This is due to the exceeding corrosion of nanotubes in high concentration electrolyte. Therefore, it is very important to reasonably control the concentration of electrolyte in the preparation of TiO_2 nanotubes arrays.

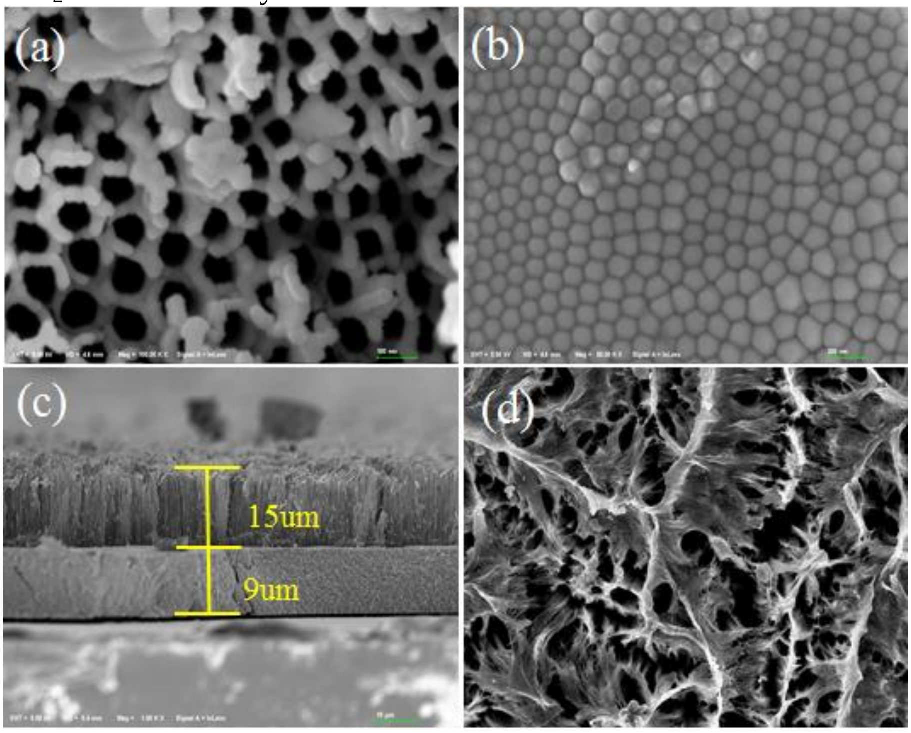

Fig.1 SEM Images of TiO_2 Nanotube Arrays in (a) Top-view, (b) Bottom-view, SEM Image of Double-layer Composite Film (c), and SEM Image of TNT-arrays with Electrolyte Concentration of 1.0wt%

Fig.2 shows the typical photocurrent density–photovoltage (J–V) curves of the DSSCs based on P25/TNT arrays double-layer with different electrolyte concentration under AM 1.5 illuminations. The detailed photovoltaic parameters are summarized in Table 1. As shown in Table 1, the conversion efficiency (η) of DSSCs can reach the maximum value of 4.20% when the concentration of electrolyte is 0.3wt%, and reach the minimum value of 1.35% when the concentration of electrolyte is 1.0wt%. There will be a wide nanotube channel appear when electrolyte concentration was high, which lead to rapid diffusion of dye molecules and electrolyte solution. However, excessively wide nanotube channel will increase the recombination rate of electrons and holes, and conversion efficiency of DSSCs decreased. Furthermore, the covering on the surface of nanotubes will block some sunlight, which also leads to the reduction of conversion efficiency. Hence, the conversion efficiency of DSSCs first increases, and then decreases, and a highest efficiency of 4.20%

was achieved at the electrolyte concentration of 0.3wt%, with short-circuit current density (Jsc) of 9.98 mAcm^{-2}, and open-circuit voltage (Voc) of 0.65 V.

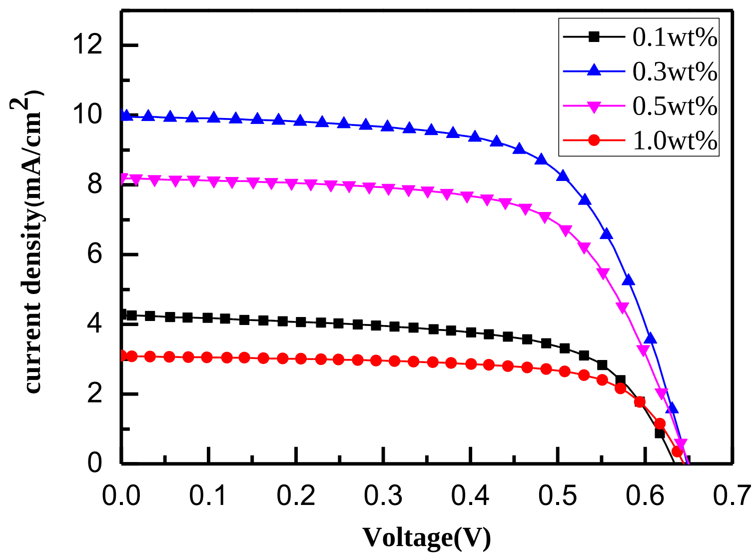

Fig.2 J–V Performance of the DSSCs Based on P25/TNT Arrays Double-layer with Different Electrolyte Concentration under AM 1.5 Illuminations

Tab.1 Comparisons of the Photovoltaic Performances of DSSCs Based on P25/TNT arrays Double-layer with Different Electrolyte Concentration

Electrolyte concentration	Jsc (mAcm^{-2})	Voc (V)	FF	η (%)
0.1wt%	4.30	0.631	0.63	1.68
0.3wt%	9.98	0.647	0.50	4.20
0.5wt%	8.21	0.649	0.64	3.45
1.0wt%	3.10	0.642	0.57	1.35

Conclusions

In conclusion, a double-layer composite TiO_2 photoanode consisting of TiO_2 nanotube arrays (TNT-arrays) as overlayer, TiO_2 nanoparticle (P25) as underlayer film was successfully fabricated and applied in DSSCs. The morphology of TNT-arrays can be controlled by appropriately changing the electrolyte concentration. Results indicate that double-layer TiO_2 DSSCs, which is fabricated from the TiO_2 nanotube arrays with the electrolyte concentration of 0.3wt% exhibits a pronounced power conversion efficiency of 4.20% under an AM1.5 G irradiation. Thus, a higher-performance DSSCs was obtained by optimizing the preparation conditions.

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