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High efficiency of dye-sensitized solar cells with two-layer mesoporous photoanodes fabricated in a low temperature process

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ABSTRACT

We fabricated, for the first time, the dye-sensitized solar cells (DSSCs) with two-layer mesoporous photoanodes in a low-temperature process, revealing a significant improvement on the performance. The lower one of the two mesoporous layers was prepared from highly crystalline, reactive anatase TiO_2 (h-TAc), which was synthesized by the hydrothermal method. Without any post treatment on the photoanodes, the power conversion efficiency (PCE) of the DSSCs incorporating h-TAc can reach as high as 6.65%, further exceeding most of the improvement without post treatment in the low temperature process reported in the literature. The enhanced PCE may be attributed to the increase of dye adsorption and the decrease of charge transfer resistance. We found the decrease of the charge interface resistance resulted from the strong chemical bonding among h-TAc particles, catalyzed by the carboxyl groups on the h-TAc surface.

1. Introduction

Solar energy is a mainly alternative energy source to fossil fuels, which can be converted into electrical power by solar cells through the photovoltaic transform mechanism. Among many kinds of solar cells, dye-sensitized solar cells (DSSCs) is one of the most promising solar cell in commercial applications because they feature simple process, easy fabrication, low cost, and high power conversion efficiency (PCE). In the past decades, the materials and interface treatments in DSSCs have been extensively studied [1–3]. The National Renewable Energy Laboratory has reported that the maximum PCE of DSSCs was confirmed to 11.9% in 2017 [4]. Recently, flexible DSSCs have received increasing attentions due to the potential in large-scale production by roll-to-roll techniques [5–9]. Because the plastic substrates and new materials for the electrodes such as silver nanowires, graphene nanosheets, and carbon nanotubes cannot withstand against high temperature, low-temperature processing is required for the flexible DSSCs [3,10–14].

Among the steps in the assembly of DSSCs, the mesoporous TiO_2 layer is sintered typically at >450 °C to eliminate organic binders and enhance the bonding strength among TiO_2 particles, reducing charge transfer resistance. To maintain the workability and adhesion of the TiO_2 paste without organic binders for the low-temperature process, Park's group [15] first reported the chemical sintering method that the viscosity of the paste and the connection of TiO_2 particles were controlled by adjusting the ratio of NH_3 to CH_3COOH (pH value). The PCE of the DSSC sintered at 150 °C by the chemical sintering method could be as high as 3.52%. The adhesion of the microporous layer on substrates could be enhanced by adding tert-butanol into the TiO_2 paste to reduce surface tension of the paste [16]. It was also found that ball-milling the TiO_2 paste could improve the strength of the microporous layer [17]. Besides the modification of the paste composition, the methods to fabricate the mesoporous layer, including electrophoretic deposition [18,19], lift-off/transfer process [20], UV-Ozone post-treatment [18,21,22], and mechanical compression method [22–24], can further enhance the PCE of DSSCs in the low-temperature process. The mechanical compression method is the most used one in the low-temperature process because of the effective increase in the connection of particles. To consider both connection and porosity, the pressure used in the mechanical compression method is required to 1000 kg/cm^2. Such high pressure significantly reduces feasibility in practical application. Moreover, most of the processing methods need extra apparatus, restrict working size, or increase fabrication cost. These problems lead to the question: are there other easier and more effective methods to improve the connection among TiO_2 particles?

In our previous study, highly reactive anatase TiO_2 chelated with

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Table 1
Properties of P25, TAc, and h-TAcs.

Samples	P25	TAc	h-TAc150	h-TAc200	h-TAc225	h-TAc250
Average particle size[a], nm	–	16.3	76.3	115.0	133.2	134.6
Crystal size[b], nm	–	4.8	7.2	10.6	13.5	15.1
BET surface area, $m^2\ g^{-1}$	50.4	288.6	206.3	121.1	88.8	70.1

[a] Determined through DLS.
[b] Calculated based on the plane (1 0 1) using Sherrer's equation.

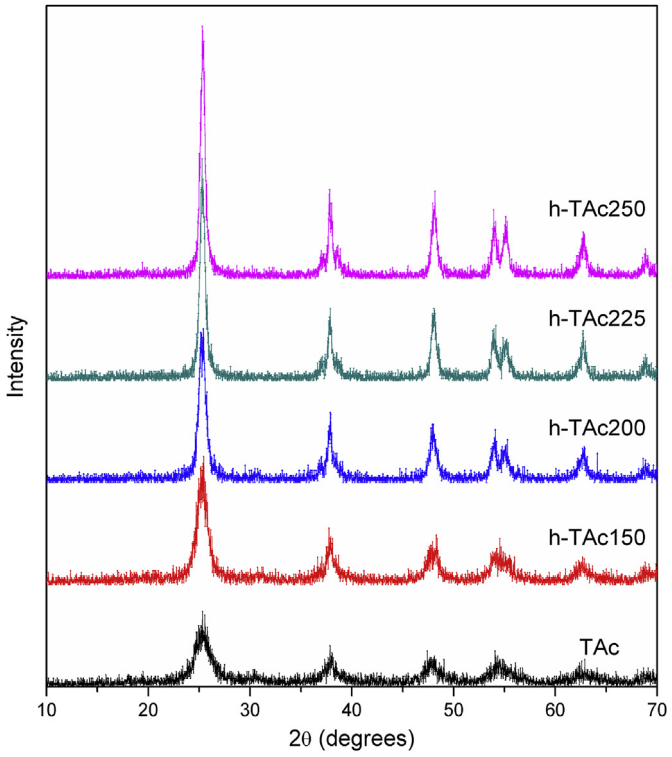

Fig. 1. XRD patterns of the TAc and h-TAcs.

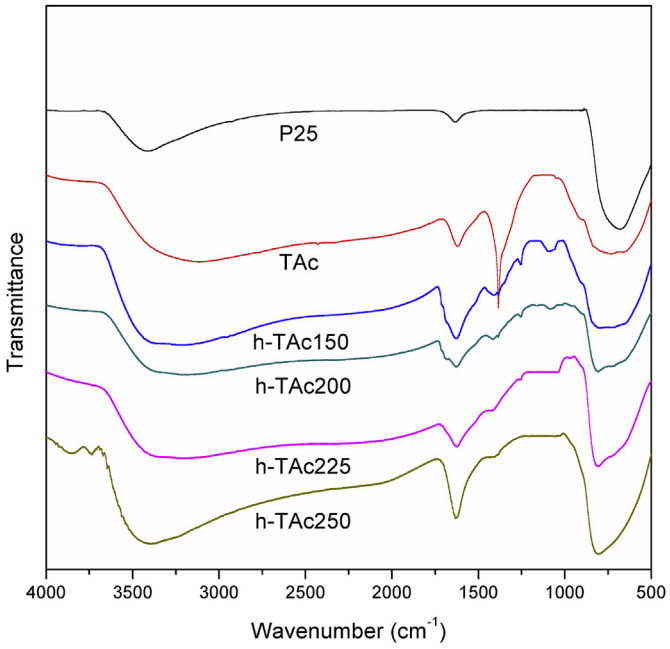

Fig. 2. FTIR spectra of the P25, TAc, and h-TAcs.

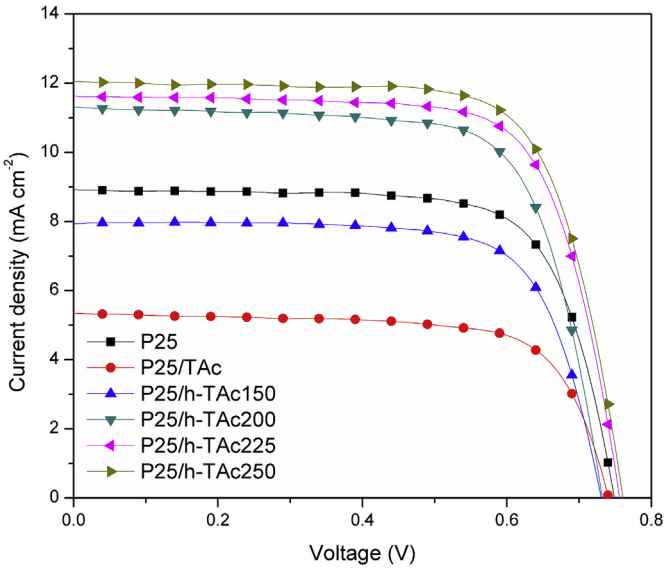

Fig. 3. Current density-voltage curves of DSSCs with various two-layer mesoporous P25/h-TAc photoanodes.

acetic acid (TAc) has been synthesized at 80 °C [25]. The TAc can be used as a sintering agent to make strong bonding at the FTO/TiO_2 and the TiO_2/TiO_2 interfaces after thermal annealing at 500 °C because the condensation reaction can be catalyzed by the carboxyl groups chelated on the surface of TAc [26]. In this study, we improved the crystallinity of TAc by the hydrothermal method (h-TAc) but still retained some carboxyl groups on its surface. Due to the highly crystalline and reactive characteristics, the h-TAc incorporating mesoporous TiO_2 photoanodes revealed good mechanical strength, much dye adsorption, and low charge transfer resistance at low-temperature process, resulting in PCE of 6.65% and 37% enhancement compared to the PCE of the pristine DSSCs, which further exceed most of improvement without post treatment reported in the literature.

2. Experimental

2.1. Preparation of h-TAc solution

TAc was synthesized using a sol-gel method, as reported previously [25]. The as-prepared TAc solution (100 ml) was added into a 225 ml vessel in a autoclave with various temperature (150, 200, 225, and 250 °C, denoted as h-TAc 150, h-TAc 200, h-TAc 225, and h-TAc 25, respectively) for 12 h. After the solution was cooled down, the h-TAc was washed three times with deionized water and then two times with EtOH through centrifugation (12000 rpm, 30 min). The purified h-TAc was re-dispersed into EtOH, resulting in h-TAc solution (3 wt%).