



## Research Paper

# Enhancing the light conversion efficiency of dye-sensitized solar cells using nanochannel TiO<sub>2</sub> prepared by spray pyrolysis



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## ABSTRACT

To enhance the photoelectric conversion efficiency of dye-sensitized solar cells (DSSCs), micron-size nanochannel TiO<sub>2</sub> particles were prepared by spray pyrolysis employing a triblock copolymer as a pore former. The as-prepared nanochannel TiO<sub>2</sub> was used to fabricate DSSC photoelectrodes, increasing the active area of TiO<sub>2</sub>/dye/electrolyte for improving charge transfer. Field emission scanning electron microscopy, transmission electron microscopy, and Brunauer-Emmett-Teller analyses were performed to determine the morphologies and characteristic properties (e.g., porosity) of TiO<sub>2</sub> particles with nanochannels obtained for various copolymer amounts. The synthesized TiO<sub>2</sub> was made into a paste and applied to DSSC photoelectrodes, resulting in a substantially increased photoconversion efficiency (7.76%) compared to that of a copolymer-free TiO<sub>2</sub>-based DSSC (3.55%). The amounts of impregnated dye in TiO<sub>2</sub>-based photoelectrode films prepared using various copolymer amounts were compared using UV-vis absorbance analysis, being substantially increased by triblock copolymer addition.

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## 1. Introduction

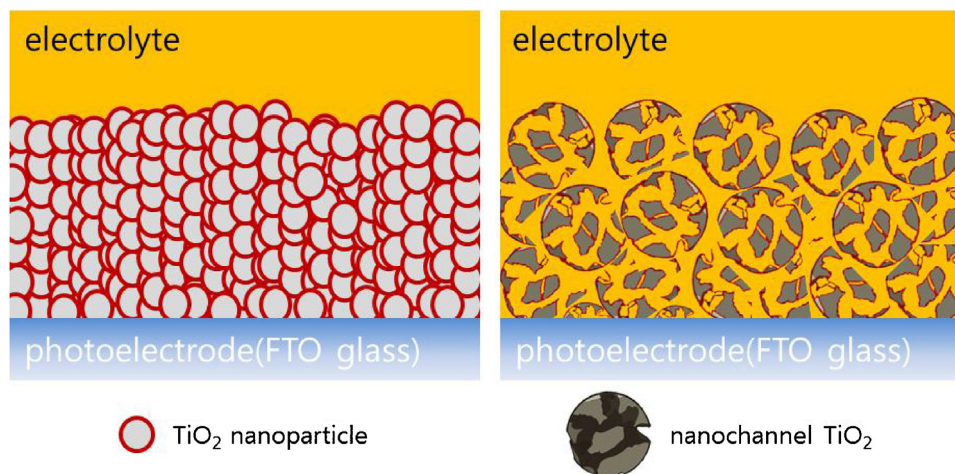
Dye-sensitized solar cells (DSSCs) are low-cost alternatives to silicon solar cells, introduced to overcome the high costs involved in raw material procurement and production [1,2]. Since then, photoelectrodes with new structures have been extensively studied to improve the photoconversion efficiency of DSSCs, which is lower than that of silicon solar cells. DSSC photoelectrodes feature a dye separating the photogenerated electrons and holes and a semiconductor oxide nanostructure transferring the generated electrons to the electrode. In particular, a variety of morphologies, such as nanoparticles [1,2], nanotubes [3–7], nanorods [8], and nanowires [9,10], have been proposed to enhance dye impregnation by increasing the TiO<sub>2</sub> photoelectrode surface area. Nevertheless, the dye impregnation enhancement achieved thus far falls short of expectations because of coagulation induced by the high surface energy of these particles. Moreover, nanoscale TiO<sub>2</sub>-based films are too dense for the facile dispersion

of liquid electrolytes, not allowing uniform photoelectrode impregnation.

Photoelectrode with porous structure can enhance dye impregnation by providing pathways for the penetration of liquid electrolyte or dye solution into the photoelectrode. Such structures also provide sites for the fast movement of large numbers of electrons and ions, owing to the increased dye/TiO<sub>2</sub>/electrolyte interface area, with similar experimental results obtained in the field of lithium secondary batteries [11]. Kang et al. [11] evaluated the performance of lithium secondary batteries after fabricating molybdenum oxide microballs with an ant-cave structure as the anode material. The nanochannel ant-cave structure was reported to improve structural stability by enabling uniform penetration of liquid electrolyte into the electrode, increasing the electrode/electrolyte interface area, facilitating charge transfer, and buffering the volume changes during the mass transfer of ions and charge-discharge cycling.

In this study, micron-scale nanochannel TiO<sub>2</sub> particles were prepared by spray pyrolysis, characterized by a number of analytical methods, and used to fabricate DSSC photoelectrodes exhibiting facilitated dye and electrolyte penetration. Fig. 1 schematically shows a photoelectrodes densely packed with nano

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**Fig. 1.** Schematic structure of a  $\text{TiO}_2$  photoelectrode packed with  $\text{TiO}_2$  nanoparticles (left) and a  $\text{TiO}_2$  photoelectrode comprising micron-scale  $\text{TiO}_2$  particles with nanochannels (right).

particles and relatively loosely packed with submicron-scale nanochannel particles.  $\text{TiO}_2$  films densely packed with nanoparticles do not have enough inter-particle space to allow liquid electrolyte or dye solutions to reach the bottom of the electrode. In contrast, porous  $\text{TiO}_2$ -based photoelectrodes comprise nanochannels acting as pathways for the even dispersion of the above solutions down to their bottom.

Micron-scale  $\text{TiO}_2$  particles with nanochannels were fabricated by spray pyrolysis, using poly(propylene glycol)-block-poly(ethylene glycol)-block-poly(propylene glycol) (triblock copolymer,  $(\text{EO})_{20}$ - $(\text{PO})_{70}$ - $(\text{EO})_{20}$ ) as a pore former, and their morphology and properties (e.g., porosity) were controlled by adjusting the block copolymer concentration. Spray pyrolysis involves spraying a precursor solution to form a droplet, solvent evaporation from the droplet within a reactor, and pyrolysis of the precipitated precursors [12–16]. Herein, a droplet was generated by spraying a  $\text{TiO}_2$  precursor solution containing the above pore former using an ultrasonic atomizer. The obtained droplet was subsequently transported by the carrier gas ( $\text{N}_2$ ) through a two-zone furnace (low-temperature drying zone and high-temperature reaction zone). In the drying zone, the solvent was gradually removed by evaporation, and the pore former within the aerosol underwent rearrangement. In the second zone, the precursor and pore former underwent decomposition and crystallization, producing micron-scale porous particles. The characteristic properties of the nano-channel  $\text{TiO}_2$  particles prepared by spray pyrolysis and cell performance of DSSC photoelectrode fabricated with as-prepared nano-channel  $\text{TiO}_2$  particles were investigated.

## 2. Experimental

### 2.1. Preparation of nanochannel $\text{TiO}_2$ particles by spray pyrolysis

Titanium isopropoxide (TTIP, Aldrich) was used as a precursor for the preparation of nanochannel  $\text{TiO}_2$  particles. Due to the occurrence of ultra-rapid hydrolysis and condensation reactions, stable chelate complexes were formed by adding acetylacetonate (acac, Aldrich) to a solution of TTIP in isopropanol (70 mL, Aldrich) to achieve an acac:TTIP molar ratio of 3:1 with subsequent stirring, whereas the precursor solution concentration was fixed at 0.3 M. The  $(\text{EO})_{20}$ - $(\text{PO})_{70}$ - $(\text{EO})_{20}$  triblock copolymer (Aldrich) used as a pore former was added to a mixture of isopropanol (50 mL),  $\text{H}_2\text{O}$  (180 mL), and 0.1 M  $\text{HNO}_3$  to obtain concentrations of 0, 15, 26, and 35 wt.% relative to the amount of TTIP, followed by stirring. The obtained copolymer solution was mixed with the precursor

solution, and the mixture was sprayed using an ultrasonic atomizer. The generated aerosol was transported by  $\text{N}_2$  carrier gas (5 L/min), passing through a 150 °C drying zone and a 600 °C reaction zone, being subsequently collected in a bag filter. The collected material was subjected to 4-h pyrolysis at 400 °C under ambient conditions to improve particle crystallinity.

### 2.2. Preparation of DSSCs with nanochannel photoelectrodes

The paste used to fabricate photoelectrode films was prepared from nanochannel  $\text{TiO}_2$  particles obtained by spray pyrolysis. A 20-mL glass vial was charged with nanochannel  $\text{TiO}_2$  (0.5 g) prepared using various copolymer amounts,  $\alpha$ -terpineol (1.5 g), ethanol (0.5 g), and ethyl cellulose (0.2 g) and mixed in a centrifugal mixer (ARM-310, THINKY Co., Tokyo, Japan) for 30 min at 2000 rpm. And then, as-prepared paste was conducted by 15-min three-roll milling for removing the air within the paste. A representative DSSC was fabricated as follows. A fluorine-doped tin oxide (FTO) glass slide ( $2 \times 1.2$  cm) was washed in acetone and ethanol (20 min each) under sonication and blown dry with nitrogen. To improve the contact between the FTO glass and  $\text{TiO}_2$  film, the former was coated with a 0.04 M  $\text{TiCl}_4$  solution, dried in an oven at 70 °C for 20 min, washed with distilled water and ethanol, and blown dry with nitrogen. The  $\text{TiO}_2$  paste prepared earlier was cast onto the  $\text{TiCl}_4$ -treated FTO glass as a 35- $\mu\text{m}$ -thick film using the doctor blade technique, followed by sintering in a box furnace at 550 °C for 30 min. The sintered photoelectrode was immersed into an ethanolic solution of the N719 dye (0.25 mM) and impregnated for 18 h in total darkness. The dye-impregnated photoelectrode was washed twice with ethanol and dried in a desiccator under nitrogen. The counter-electrode was fabricated by casting Pt paste onto FTO glass using the doctor blade technique, followed by 20-min sintering in a box furnace at 400 °C. The photoelectrode and counter-electrode layers prepared as described above (with a 60- $\mu\text{m}$ -thick Surlyn layer sandwiched between them) were joined by annealing on a heating plate (180 °C). Finally, the electrolyte was injected through the hole between the sealed electrodes, and the fabrication process was completed by sealing the hole with a soldering iron using Surlyn and a cover glass.

### 2.3. Characterization

The morphologies of nanochannel  $\text{TiO}_2$  particles prepared by spray pyrolysis were investigated by field emission scanning electron microscopy (FE-SEM; SU8010, HITACHI) and transmission

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