Manufacture of dye-sensitized solar cells using ultrasonically treated TiO₂

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1. Introduction

Efforts to improve the performance of dyesensitized solar cells (DSSC) have led to innovative strategies such as utilizing sonication to enhance the properties of titanium dioxide (TiO₂) particles used in the fabrication of DSSC photoelectrodes.^{1,2)} This study investigates the effects of different sonication treatments on TiO₂ particles and their subsequent incorporation into DSSC photoelectrodes to improve conversion efficiency.^{3,4)} Sonication energy techniques create defects on the surface of TiO₂ particles and result in a corresponding increase in surface area. This increased the amount of dye adsorption and resulted in an approximately 20% increase in the energy conversion efficiency of the DSSC.

2. Materials and Methods

The DSSC fabricated in this experiment were made using TiO₂ (anatase 99.9%, US Research Nanomaterials, Houston, TX), α -terpineol (98.5%, Samchun Chemicals, Seoul, South Korea), chloroplatinum hydrate (Sigma Aldrich, St. Louis, MO), N719 dye (Solaronix, Aubonne, Swiss), ethanol (HPLC grade, Duksan Co., Ansan, South Korea), and ethylcellulose 10 cP (extra pure, Daejung Chemicals & Metals, Siheung, South Korea). Sonication equipment included an ultrasonic cleaner (JAC2010, KODO Technical Research Co., Hwasung-si, South Korea) and an ultrasonic horn (VC750, Sonics & Materials, Newtown, CT).

The TiO₂ paste was prepared by a sol-gel method in which 25 ml of ethanol, 2.15 ml of α -terpineol, and 0.6 ml of distilled water were added to 2 g of TiO₂ and stirred at 300 rpm for 15 min at 120 °C. Then, 0.3 g of ethyl cellulose was added and stirred until the paste was complete. The finished paste was applied to the FTO plate by a doctor blade method, and TiO₂ photoelectrodes were prepared by a process of calcination at 450 °C for 2 h at a temperature rise rate of 5 °C/min. The photoelectrodes were immersed in N719 dye (0.5 mM) in ethanol for 24 hours at room temperature.

For the preparation of the ultrasonic cleaner- TiO_2 paste, we added an indirect sonication process



Fig. 1 Schematic diagram of the process for fabricating dye-sensitized solar cells using cleaner-ultrasonic and horn-ultrasonic sonicated TiO_2 and pure TiO_2 .

to the same method for TiO_2 paste preparation. The ultrasonic cleaner- TiO_2 solution was prepared by adding 2 g of TiO_2 to 50 ml of ethanol, followed by indirect sonication (high intensity) in an ultrasonic cleaner bath for 30 minutes. The ultrasonic cleaner- TiO_2 photoelectrode was then prepared in the same way as the TiO_2 .

The ultrasonic horn-TiO₂ paste was prepared in the same way as the ultrasonic cleaner-TiO₂ paste. The ultrasonic horn-TiO₂ solution was prepared by directly sonicating a solution of 2 g of TiO₂ in 50 ml of ethanol with an ultrasonic horn for 30 min. The ultrasonic horn-TiO₂ photoelectrode was then prepared in the same manner as the TiO₂.

Fabricate the counter electrode by drilling two holes in the FTO glass to inject the electrolyte. The counter electrode was prepared by coating the FTO glass with H₂PtCl₆ solution by spin coating and then calcining it at 350° C for 1 hour. After combining the prepared counter electrode with the working cathode, iodine solution AN-50 was injected as the electrolyte and the holes were closed to complete the DSSC (**Fig. 1**).

3. Result and discussion

Two methods were employed for TiO_2 particle treatment: direct ultrasonication using an ultrasonic horn and indirect treatment through an ultrasonic cleaner. The TiO_2 particles (Fig. 2 a) are

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agglomerated, while the sonicated ultrasonic cleaner-TiO₂ and ultrasonic horn-TiO₂ particles are dispersed. The dispersion of ultrasonic horn-TiO₂ was relatively higher than that of ultrasonic cleaner-TiO₂ (**Fig. 2** b and c), which is consistent with the findings that ultrasound affects particle dispersion.⁵) **Fig. 2** (e and f) shows that the roughness on the outside of the particles increased due to sonication. The cavitation bubbles generated by sonication of ethanol mixed with TiO₂ contributed to the formation of defects on the surface of the particles, which contributed to the activation of the particle surface, which enhanced the photocatalytic performance.



Fig. 2 (a-c) SEM images of (a) Pure TiO₂; (b) ultrasonic cleaner- TiO₂; (c) ultrasonic horn-TiO₂. And (d-f) TEM images of (d) Pure TiO₂; (e) ultrasonic cleaner-TiO₂; (f) ultrasonic horn-TiO₂

The semicircles in **Fig. 3** represent the charge transfer resistance measured at about 15 k Ω , with pure TiO₂ having the highest. The semicircles for the ultrasonic cleaner-TiO₂ and the ultrasonic horn-TiO₂ are around 7-8 k Ω , showing a charge transfer resistance that is 50% lower than TiO₂.



Fig. 3 EIS Nyquist plot (under dark condition) of (a) Pure TiO_2 ; (b) ultrasonic cleaner- TiO_2 ; (c) ultrasonic horn- TiO_2 .

Fig. 4 shows a J-V graph measuring energy conversion efficiency. The ultrasonic cleaner-TiO₂ had an energy conversion efficiency of 2.82%, an improvement of about 20% over TiO₂ (2.35%). ultrasonic horn-TiO₂ showed an efficiency of 3.35%, which was 43% higher than TiO₂ (2.35%) and about 19% higher than ultrasonic cleaner-TiO₂.

In conclusion, sonication contributed to

increasing the surface activity of the particles, decreasing the electron transfer resistance, and increasing the energy conversion efficiency. These findings highlight the innovative potential of sonication in DSSC technology and provide new directions for efficient and practical DSSC research.



Fig. 4 Photocurrent density–voltage (J-V) graph of the DSSC (under one-sun illumination of 1000 mW/cm^2). (a) Pure TiO₂; (b) ultrasonic cleaner-TiO₂; (c) ultrasonic horn-TiO₂.

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