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# Influence of dye-concentration on the light-scattering effect in dye-sensitized solar cell

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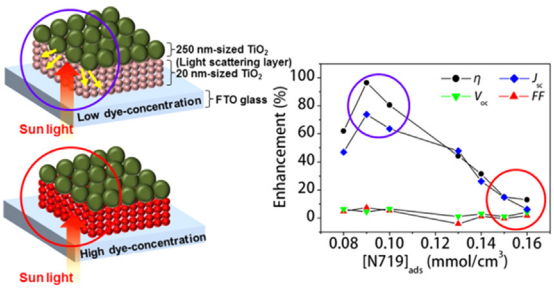
## HIGHLIGHTS

- The role of scattering layer for DSSC was investigated.
- The conversion efficiency of DSSC was affected by the scattering layer.
- The efficiency of DSSC significantly was attributed to the dye concentration adsorbed on $TiO_2$.
- The effects of scattering layer were discussed in terms of photo-dynamic results.

## GRAPHICAL ABSTRACT

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## ABSTRACT

We investigate the influences of dye-concentration for scattering layer in a dye-sensitized solar cell (DSSC), which is consisted with N719 dye on a bilayer anode with 250 nm-sized $TiO_2$ film onto 20 nm-sized $TiO_2$ film, and compared to a monolayer anode 20 nm-sized $TiO_2$ film. The photovoltaic conversion efficiency ($\eta$) of DSSC with the bilayer anode including a light-scattering layer (LSL-DSSC) was enhanced markedly compared to that of the DSSC with normal transparent layer anode (NTL-DSSC). This implies that the light-scattering layer increased the light amount absorbed by dyes. The $\eta$ was improved more than 80% in low concentration of dye adsorbed on $TiO_2$, which is caused by the penetrating light up to the scattering layer. The scattering effect is more effective in lower concentrations of dye. The photovoltaic performance of LSL-DSSC was characterized by the electrochemical and photophysical analysis comparing with the NTL-DSSC. Moreover, the charge recombination and dye regeneration dynamics for DSSC were investigated using the transient absorption spectroscopic measurements.

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## 1. Introduction

Since the development of dye-sensitized solar cells (DSSCs) by O'Regan and Grätzel in 1991 [1], DSSC technology has been regarded as one of the promising clean photovoltaic technologies due to their low cost and relatively high efficiency [2–5]. Although numerous oxide systems have been tested in order to obtain efficient light-to-electric conversion in DSSCs [6,7], mesoporous crystalline $TiO_2$ is the most convincing material for DSSC photoanodes [8]. A lot of research has been carried out to improve the performance of DSSC by modification of various components of DSSC [9–18]. One effective method to increase the light harvesting capability is introducing optical scatters into the photoanode [19,20]. Typically, bilayered $TiO_2$ structure, consisting of a main $TiO_2$ layer prepared from the tiny nanoparticles (<few tens nm) and a

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light-scattering layer from the large particles (>few hundreds nm), have been designed for efficient utilization of the solar spectrum and enhancement of photovoltaic conversion efficiency ($\eta$) [21–30].

The light-scattering layer on top of the main $TiO_2$ layer confines the incident light within an electrode or diffracts it backward. Thereby, the dye molecules anchored on the $TiO_2$ surface utilize solar spectrum more efficiently. Especially, the quantum efficiency in the longer wavelength region can be enhanced relatively more. There were some suggestions to explain the scattering effects. First, the particles showed strong dependency on their diameter and refractive index [31]. Second, regularly ordered arrays of monodispersed $TiO_2$ spheres can generate photonic reflection peaks, the positions of which also correlate with their diameters [32]. Therefore, the large particles of rutile [33], $ZrO_2$ [34], $SrTiO_3$ [35] or other high-refractive-index metal oxides [36–38] have been used as scattering layer for DSSC [39,40]. On the other hand, the DSSC including spherical Ag nanoparticles show the highest solar conversion efficiency, which is attributed to the enhanced plasmonic effect and scattering mechanism in the cell [41]. The photovoltaic performance also can be improved by using the modified film such as uniform crack-free film [42].

In this work, we prepared normal DSSC device using 20 nm-$TiO_2$ particles in the average diameter. Also, 250 nm-$TiO_2$ particles were used as the light-scattering layer for the incident light. In order to elucidate the penetrating behaviors, their photo- and electrochemical properties were examined under the differently adsorbed dye-concentrations. We have attempted to understand the influence of concentration effects for the scattering layer in DSSCs.

## 2. Experimental

### 2.1. Preparation and characterization of the photoelectrode

A fluorine-doped tin–dioxide (FTO) conducting glass (TEC-8, Pilkington Co.) substrate was cleaned in acetone using an ultrasonic bath for 15 min, then rinsed with ethanol and water, and finally dried in an oven at 80 °C for 1 h. A mask with an open area of 0.6 cm × 0.6 cm was placed on the FTO glass for photoelectrode deposition. The 20-nm-sized $TiO_2$ paste was prepared according to procedures reported previously [43,44], and placed in the mask opening for screen printing (SM-S320, Sun Mechanix Co.). The thickness of the $TiO_2$ film was controlled by repeated cycles of screen printing followed by drying at 120 °C for 5 min. The light-scattering layer with $TiO_2$ particles of ca. 250 nm in diameter (STP-250N, ENB Korea Co.) was coated on top of the main $TiO_2$ film. Whenever the $TiO_2$ film was coated by screen printing, the coated $TiO_2$ film was kept in a clean box saturated with ethanol for approximately 10 min to reduce the surface irregularities of the coated film (i.e., leveling). Then, the $TiO_2$ film was completed with sintering at 500 °C for 30 min to remove the viscous solvent and impurities. The $TiO_2$ film was then soaked for 24 h at room temperature in a 0.01–0.5 mM N719 dye solution (Solaronix Co., in absolute ethanol) to control the concentration of adsorbed dye. After preparing the photoelectrode, un-adsorbed dye molecules were rinsed with absolute ethanol.

Two pin-holes for injecting the electrolyte were drilled into the rinsed FTO glass using a drilling machine (Dremel Kit 335, Bosch Co.). A Pt counter electrode was prepared by depositing a Pt solution (0.05 M $H_2PtCl_6$, Aldrich Co., in absolute ethanol) onto the FTO glass via spin coating (ACE 200, i-Nexus Inc.) and annealing at 400 °C for 30 min in air. The prepared photoelectrode and counter electrode were sealed with a hot-melt sealing sheet (SX 1170-100, Solaronix Co.) with a thickness of 100 μm. The DSSC was completed by injecting a redox electrolyte consisting of 0.6 M PM II (1-methyl-3-propylimidazolium iodide, Solaronix Co.), 0.03 M $I_2$ (Aldrich Co.), 0.1 M guanidinium thiocyanate (Aldrich Co.), and 0.5 M 4-tert-butylpyridine (Aldrich Co.) in a mixture of acetonitrile and valeronitrile ($v/v$ = 85:15, Aldrich Co.).

The $TiO_2$ particles in the photoelectrode were characterized via field-emission scanning electron microscopy (SEM) (S-4160, Hitachi Co.). The $TiO_2$ film thickness was measured using a stylus profiler (Alpha-Step IQ, KLA-Tencor Co.). The concentration of adsorbed N719 dye on the $TiO_2$ film was calculated using UV/Vis spectrometry (UV-160A, Shimadzu Co.) [43,44].

### 2.2. Measurement of photoelectricity in the DSSC

The current density–voltage ($J$–$V$) characteristics were measured using a source meter under white-light irradiation from a 450-W xenon lamp (Thermo Oriel Instruments). Prior to measurement, the light intensity was calibrated using a Si reference diode equipped with an air mass (AM) 1.5-G filter (KG5). The incident photon–to–current efficiency (IPCE) spectra were recorded as a function of wavelength from 300 to 900 nm using an IPCE measurement apparatus (QEX7, PV Measurements Inc.). Electrochemical impedance spectroscopy measurements were performed using an electrochemical workstation (Versastat4, Princeton Applied Research) in frequency range of 100 kHz to 100 mHz with amplitudes of 10 mV over the $V_{oc}$.

### 2.3. Measurement of photodynamics in the DSSC

The nanosecond transient absorption measurements were conducted using laser flash photolysis [44,45]. The DSSCs were excited by nano-second laser pulses produced by an optical parametric oscillator (Continuum, Surelite OPO plus), pumped by third-harmonic generation (355 nm, FWHM of 4.5 ns) from a Q-switched Nd:YAG laser (Continuum, Surelite II-10). The excitation pulses (540 nm, 10-Hz repetition rate) were attenuated by neutral filters to reduce the pulse intensity (<30 μJ/cm$^2$) on the sample. Light from a xenon arc lamp (ILC Technology, PS 300-1) was passed through a monochromator (or a band-pass filter with a FWHM of ~20 nm) and focused on the sample for the transient absorption measurements. Temporal profiles were measured with a monochromator (Dong-Woo Optron, Monora 500i) equipped with a fast photomultiplier tube (Zolix Instruments Co., CR 131) and a digital oscilloscope (Tektronix, TDS-784D). The reported signals were averaged over 3000 events to obtain satisfactory signal-to-noise ratios. The transient absorption spectra were measured with an intensified charge-coupled device (Ando, iStar).

## 3. Results and discussion

### 3.1. Light penetration behaviors in NTL-DSSC and LSL-DSSC

In fabricating the DSSCs, the main $TiO_2$ layer was formed by applying ~20–nm-wide nanoparticles on the FTO substrate using the screen printing method. $TiO_2$ particles 250 nm in diameter were then overlay-coated using the same coating method, which can scatter the visible light. The surface and cross-sectional structure of the photoanode were characterized using SEM as shown in Fig. 1. The cross-sections showed the bilayer structure, consisting of the main layer with nanocrystalline $TiO_2$ and the scattering layer. The thicknesses of the NTL and LSL were 15 and 7.5 μm, respectively, as shown in Fig. 1(a). The scattering layer with large-sized $TiO_2$ is expected to reduce the dye loading capacity as a result of the reduced surface area compared to that of the smaller-sized $TiO_2$. Therefore, we could visually identify the lesser adsorption of dye on the scattering layer (the white color, Fig. 1(b)) compared with that

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