



Ytterbium oxide nanodots via block copolymer self-assembly and their efficacy to dye-sensitized solar cells

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ABSTRACT

In this study, we develop a novel phosphor, Yb₂O₃, to be used as the spectral converter in dye-sensitized solar cells (DSSCs) for the efficient capture of ultraviolet light via down-conversion. These zero-dimensional nanodots with a high refractive index also allow more light to be trapped and can prevent charge recombination at the interfaces in the DSSCs. Compared to DSSCs without the nanodots, the DSSCs fabricated with the Yb₂O₃ nanodots exhibits higher power-conversion efficiencies for both the N719 (10.5%) and CSD-01 (20.5%) dyes. The multifunctionality of the Yb₂O₃ nanodots provides a new route for improving the performance of DSSCs.

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1. Introduction

To aid the efforts of those researching clean and sustainable energy sources against a backdrop of the impending depletion of fossil fuels and the global warming crisis, photovoltaic devices have been extensively studied across the world. Since O'Regan and Grätzel invented a dye-sensitized solar cell (DSSC) in 1991 [1], it has attracted much attention because of its distinctive features, such as its color and transparency, low weight, flexibility, workability under low light conditions, and easy integration with buildings and automobiles [2,3]. The DSSC is a photoelectrochemical device that mimics the principles of natural photosynthesis, and its photocurrent is generated at the interface between the photosensitized semiconductors and redox electrolytes under solar illumination. To date, the many attempts to enhance its performance have mainly been focused on developing photosensitizers with a wider spectral absorption band and higher molar extinction coefficients [4–6]. Another interesting approach is applying efficient photon management to increase the light harvesting efficiency (LHE) of the cell. Most of this work has focused on increasing the optical path length or local electromagnetic energy within the absorbing electrode [7–10]. However, the harvesting of ultraviolet (UV) and near-infrared (NIR) light is still difficult because most of the

photosensitizers used in DSSCs only absorb light with wavelengths ranging from 400 to 800 nm. Recently, to maximize the usage of solar photons, luminescent materials based on trivalent lanthanide ions have been considered as spectral converters because of their rich energy-level structure (known as the Dieke diagram) [11]. For example, Wu et al. demonstrated that the rare-earth compound YF₃:Eu³⁺ could be used as a UV-absorbing spectral converter to improve the DSSC performance via down-conversion, in which one UV photon is transformed into two visible photons [12]. On the other hand, Zhang et al. reported NaYF₄:Yb³⁺,Er³⁺ nanoparticles that can improve the DSSC performance via up-conversion, which extends the spectral response range to the infrared region [13]. Despite recent progress in preparing such fluorides, the synthesis of high-quality nanocrystals still requires complicated processes and high reaction temperatures.

Recently, arrays of zero-dimensional nanoparticles with high refractive indices have been adopted to confine light in the propagation direction, and thus, such nanoparticles can be employed to trap the incident sunlight in a solar cell [14]. To prepare self-ordered nanomaterials on solid substrates, block copolymers (BCPs), which consist of two or more chemically different polymers covalently connected, have been extensively exploited [15–17]. In particular, BCP micelles, in which a solvophilic block forms a swollen corona around a solvophobic core, have been utilized as templates for inorganic nanomaterials, including metals, semiconductors, and ceramics. The precursors of such inorganic materials are incorporated into the micellar core, then the micellar films are

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formed on the substrate by either spin coating or dip coating, and finally the BCP templates are removed with UV exposure, O₂ plasma etching, or calcination. For example, Lohmueller et al. fabricated hexagonally ordered metal nanoparticles (Au, Pt, and Pd) on glass substrates by using polystyrene-block-poly(2-vinylpyridine) (PS-*b*-P2VP) micelles [18]. Anthony et al. reported the fabrication of two-dimensional arrays of luminescent metal-chalcogenide nanocrystals [19]. In addition, Kim et al. demonstrated the synthesis of TiO₂ and ferroelectric PbTiO₃ nanodot arrays on platinumized silicon substrates by using polystyrene-block-poly(ethylene oxide) (PS-*b*-PEO) loaded with sol-gel precursors [20].

In this study, we developed a novel phosphor, Yb₂O₃, to be used as the spectral converter in DSSCs for the efficient capture of the full solar spectrum. We found that the Yb₂O₃ nanodots fabricated via the BCP self-assembly can trap more light and prevent charge recombination at the interfaces in the DSSCs.

2. Experimental methods

2.1. Preparation of ytterbium oxide nanodots

All of the chemicals were purchased from Sigma Aldrich and used without further purification. The poly(styrene-block-4-vinylpyridine) diblock copolymer (PS-*b*-P4VP, $M_{n,PS} = 41$ kg/mol, $M_{n,P4VP} = 24$ kg/mol, $M_w/M_n = 1.06$) was obtained from Polymer Source Inc. First, the PS-*b*-P4VP was dissolved in toluene at 80 °C and stirred for 2 h to yield a solution with a concentration of 0.5 wt.%. The Yb precursor solution (1.0 wt.%) was prepared by dissolving ytterbium(III) chloride hexahydrate (YbCl₃·6H₂O) in ethanol. This solution was added to the BCP solution at different molar ratios of YbCl₃·6H₂O to 4VP (0.1, 0.3, and 0.5), and then the mixture was vigorously stirred for a week. The precursor-loaded inverse micelles were spin-coated onto transparent fluorine-doped SnO₂ (FTO)-coated conducting glass (TEC 8, Pilkington, 2.2-mm-thick, sheet resistance = 8 Ω/□) at 2000 rpm for 60 s. The polymer templates were eliminated with O₂ plasma etching at 80 W for 7 min, leaving the zero-dimensional ytterbium oxide nanodots on the substrate.

2.2. Characterization of the Yb₂O₃ nanodots

Non-contact atomic force microscopy (AFM, XE-120, Park Systems, Korea) was used to image the surface morphology of the prepared nanodots. Al-coated non-contact tips (Nanosensors, PPP-NCHR, Switzerland, force constant (k) = 42 N/m, resonance frequency = 330 kHz) were used for the AFM imaging, and the scan rate was 1.0 Hz. The optical transmittance spectra of the nanodots were collected with a UV/VIS/NIR spectrophotometer (V-670, JASCO, USA) over a wavelength range of 300–1200 nm. The excitation and photoluminescence (PL) spectra were measured with a photoluminescence spectrometer (iHR320, HORIBA Ltd., France); the excitation wavelength was 350 nm. X-ray photoelectron spectroscopy (XPS) was performed with a K-alpha XPS system (Thermo Fisher Scientific Inc., UK) using a monochromated Al Kα X-ray source with an energy of 1486.6 eV. The spectra of the Yb 4d and O 1s energy levels were calibrated with respect to the C 1s peak at 285.0 eV of the adventitious carbon that had formed on the substrate's surface. The oxide nanodots used for the optical measurements and XPS analysis were prepared with an YbCl₃·6H₂O/4VP molar ratio of 0.3 and annealed under the same conditions used for the photoanode fabrication.

2.3. DSSC fabrication and characterization

TiO₂ photoanodes were screen-printed on ytterbium oxide nanodot arrays, which were formed on FTO:glass substrates. TiO₂

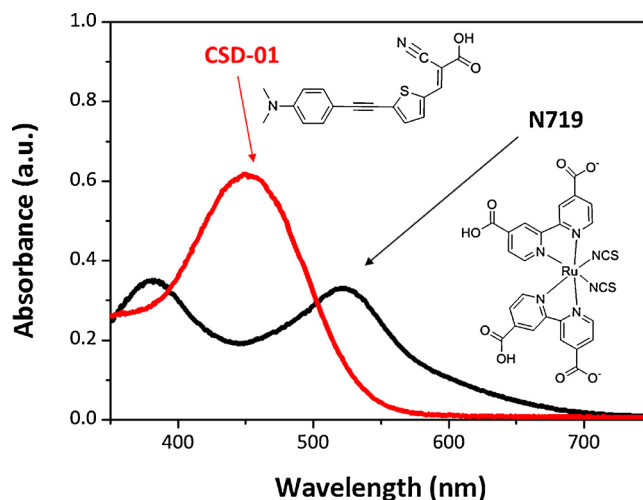


Fig. 1. Chemical structures and UV-vis absorbance spectra of the N719 (black spectrum) and CSD-01 (red spectrum) sensitizer molecules. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of the article.)

photoanodes on bare FTO:glass substrates were also fabricated for a control experiment. The resulting layers were dried for 10 min at 300 °C on a preheated hot plate. The screen printing and drying processes were repeated until a thickness of approximately 20 μm was obtained. Subsequently, the thick TiO₂ films were placed in a muffle furnace and gradually heated to 300 °C over a 30 min period, heated at 300 °C for 1 h, heated to 575 °C over a 30 min period, sintered at 575 °C for 1 h, and then cooled to room temperature. The active area of the electrodes was 0.25 cm². The prepared photoanodes were immersed in a 0.04 M solution of TiCl₄ at 75 °C for 1 h, rinsed with deionized water, and then sintered at 500 °C for 30 min. They were exposed to O₂ plasma for 10 min and then immersed for 24 h in one of two photosensitizer-containing ethanol solutions (0.5 mM) (either cis-diisothiocyanato-bis(2,2-bipyridyl-4,4'-dicarboxylato)ruthenium(II) bis-(tetrabutylammonium), N719, Solaronix, Switzerland, or 2-cyano-3-(5-((4-(dimethylamino)phenyl)ethynyl)thiophen-2-yl)acrylic acid, CSD-01, Sooyang Chemtec, Korea) [21]. Fig. 1 shows the UV-vis spectra of the N719 and CSD-01 dyes recorded in *N,N*-dimethylformamide (DMF, 1 × 10⁻⁵ M). The Pt counter electrodes were prepared on the FTO-coated glass with magnetron sputtering after two holes were drilled in the glass. Both the dye-sensitized TiO₂ electrode and Pt counter electrode were sealed with a 60-μm-thick layer of Surlyn (Solaronix, Switzerland). An ionic liquid electrolyte (0.60 M of butyl methyl imidazolium iodide, 0.03 M of I₂, 0.50 M of 4-tert-butylpyridine, and 0.1 M of guanidinium thiocyanate in an 85:15 (v/v) mixture of acetonitrile:valeronitrile) was injected into the rear side of the counter electrode. The photovoltaic characteristics of the devices under AM 1.5 illumination (equivalent to one sun, 1 kW/m²) were investigated with a solar cell current-voltage (*I*-*V*) measurement system (K3000 LAB, McScience, Korea). The photocurrent density (J_{sc}), open-circuit voltage (V_{oc}), fill factor (FF), and power-conversion efficiency (η) were simultaneously measured.

3. Results and discussion

3.1. Self-ordered ytterbium oxide nanodots

Homogeneous spherical micelles were formed by using a 0.5 wt.% solution of the amphiphilic PS-*b*-P4VP copolymer in toluene. The Yb precursor was incorporated into the P4VP cores

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