



Metal-free organic-dye-based flexible dye-sensitized solar textiles with panchromatic effect



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ABSTRACT

The first completely solid, multi-color, flexible solar textile was fabricated for use as a metal-free organic dye-sensitized solar textile with a satin structure. A one-dimensional, ZnO nanorod directly grown on a fiber-type stainless steel wire was selectively sensitized using three different indoline dyes (D102, D131 and D149) with different optical properties for the photoanode, and a Pt-coated wire was used as the counter electrode. A solar textile composed of five photoanodes and counter electrodes with three different main color was woven into a satin structure and casted with a solid electrolyte. For multi-color solar textiles, both the efficiency and current density were higher than the single textiles with the same main color, indicating the panchromatic effect of a multi-color system. The lifetime of each dye loaded sample was evaluated under accelerated conditions and further compared with the color values of the Commission International d'Eclairage LAB.

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1. Introduction

Dye-sensitized solar cells (DSSCs) are promising candidates for next-generation photovoltaic devices due to their advantages of easy production, low cost, flexibility and transparency compared to conventional solar cells [1–7]. However, the highest efficiency of 12.1% has been achieved using ruthenium complexes (Ru metal dye) – TiO₂ DSSCs [8] with a relatively thick TiO₂ layer (minimum: 10 μm), which is required to absorb a significant fraction of the incident light [9]. This requirement limits the flexibility and increases the rigidity of the entire system. The major factor responsible for this limitation is the rare and noble Ru metal sensitizer in this system, which is the most commonly used dye in DSSCs and has a low extinction coefficient at visible light wavelengths [10–13]. Therefore, metal-free organic dyes with high molar extinction coefficients that allow for complete light absorption at important wavelengths are gaining interest in DSSC research [14,15]. Organic-dye-based DSSCs can be also constructed as thin films and have the additional advantage of a comparably low cost for synthesis and purification [15,16]. Moreover, the tunable molecular design of organic sensitizers leads to more applications of

the final devices because it allows changes in the absorption and the electrochemical and color properties [11,15].

Among the various types of organic dyes for DSSCs, indoline dye based DSSCs have recently achieved the highest conversion efficiency of 9% under full sunlight [17]. The indoline dyes are common dyes with different optical properties that depend on their molecular designs, such as cyanoacrylic acid (D131, yellow), single (D102, red) and double rhodanine acetic acid (D149, purple hue) [18,19], which is an attractive factor for further applications of DSSCs [20]. However, in spite of the high absorbance coefficient values, organic-dye-based DSSCs demonstrate relatively low performance compared to Ru systems due to their sharp and narrow absorption spectra [7,15,21]. Various methods, such as multi-color arrangements [22,23], dye cocktails and co-sensitization [24,25], have been proposed to expand the light absorption spectra of the organic-dye-based DSSC systems. In an attempt to avoid unfavorable interactions between the dye molecules, an accurate positioning method [26] and a simple fabrication using a desorption and adsorption method [27] were recently demonstrated. However, during the desorption-adsorption process, these methods still include the possibility of damaging the dye-loading site, which involves an elaborate process to control the desired thickness at the specific site. Therefore, an advance in research resulting in the accurate and specific positioning of each dye, which combines a simple and cost-efficient fabrication method, is required for multi-dye DSSCs

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systems. Moreover, a compromise between the efficiency of the energy generation and the color fastness of the dyes (for esthetic purposes) should be considered for further applications.

To satisfy the energy demands in a broader range of applications, flexible DSSCs have greatly enriched the possibility of applications with increased mobility and adaptability [28–35]. Advanced types of flexible DSSCs with lightweight, foldable and cylindrical fiber-shaped electrode materials are being introduced for fiber or textile geometries [31–35]. This type of flexible DSSCs is a novel idea that fulfills the requirements for practical applications in terms of roll-to-roll and increased size production by lengthening or widening the cell. However, because most of these products are only suggested possibilities, more research combined with a proper fabrication method is required to introduce flexible DSSCs as real products. Furthermore, the limitations due to the relatively weak and unstable binding arrangements of the electrodes and the requirements for appropriate morphological stability and electrical properties of the semiconducting layer and a stable electrolyte that is resistant to oxidation, leaking or evaporation are topics that should be considered.

Herein, we report completely solid, flexible, dye-sensitized solar textile (DSST) that were woven into the satin structure of a conventional textile. Following the previous work [36], the five respective photoanodes in our base structure will provide selective loading sites for three different indoline dyes for the multi-color DSST. By considering the durability of the DSST, zinc oxide nanorods (ZnO NR) directly grown on stainless steel (SS) wire substrates and a solid polymer electrolyte consisting of a poly(vinyl chloride)-*g*-poly(oxyethylene methacrylate) (PVC-*g*-POEM) graft copolymer were selected for this study. The characteristics of the ZnO NR grown photoanode were determined using scanning electron microscopy (SEM), transmission electron microscope (TEM) and X-ray diffraction (XRD). The multi-color effect and corresponding energy conversion efficiencies were evaluated using UV–visible spectroscopy and a solar simulator with different ratios of the three types of indoline dyes in a single and multi DSST system. The photodegradation and the color fastness of each organic sensitizer on the ZnO NRs was further demonstrated using UV–visible spectroscopy and Commission International d'Eclairage (CIE) LAB color scale after exposure to accelerated conditions with different irradiation times.

2. Experimental section

2.1. Materials

Fiber-type conductive SS wires with 0.1-mm diameters (Duckheung, Korea) were selected as the photoanode and counter electrode base materials. Poly(vinyl chloride) (PVC, $M_w = 97,000$ g/mol, $M_n = 55,000$ g/mol), poly(oxyethylene methacrylate) (POEM, $M_n = 475$ g/mol), 1-methyl-3-propylimidazolium iodide (MPII), lithium iodide (LiI), iodine (I_2), and copper (I) chloride (CuCl, 99%), diethylzinc solution (1.0 M in hexane), zinc nitrate hexahydrate, hexamethylenetetramine (HMTA), poly(ethylene imine) (PEI, end-capped, molecular weight 800 g mol⁻¹), ammonium hydroxide solution, titanium diisopropoxide bis(acetylacetonate) and chloroplatinic acid hexahydrate ($H_2PtCl_6 \cdot 6H_2O$) were obtained from Aldrich, and methanol, isopropyl alcohol (IPA), 1-methyl-2-pyrrolidinone (NMP), tetrahydrofuran (THF) and acetonitrile were obtained from JT Baker. Poly(ethylene terephthalate) (PET) film of 25 μ m thickness was purchased from SKC (Korea) and the three types of indoline dyes, namely C₃₇H₃₀N₂O₃S₂ (D102), C₃₅H₂₈N₂O₂ (D131), and C₄₂H₃₅N₃O₄S₃ (D149), were purchased from Mitsubishi Paper Mills (Japan). All of the solvents and chemicals were of reagent grade and used as received without further purification.

2.2. ZnO NR on SS wires (ZnO NR-SS) for the photoanodes

Scheme 1(a) shows the growing process of the ZnO NR on SS to obtain the photoanode. The SS was purified via 30 min sonication in IPA (100 ml) and was spin-coated with a ZnO seed solution for 30 s, which was prepared by dissolving 0.37 ml of diethylzinc solution in 5 ml of THF with 1 h stirring at room temperature. After the thermal decomposition process at 450 °C for 30 min, the seeded wires were stored inside a glass bottle filled with an aqueous growing solution at 90 °C in an oven for 5 h. Zinc nitrate hydrate (0.050 M), HMTA (0.050 M), PEI (0.005 M) and ammonium hydroxide (0.35 M) was homogeneously mixed for the growing solution. After rinsing the wires with deionized water, the wires were sintered at 450 °C for 30 min.

2.3. Pt-coated counter electrodes

The SS wires were spin-coated with a Pt solution containing 0.35 g of chloroplatinic acid hexahydrate (H_2PtCl_6) dissolved in 100 ml of IPA for 30 s. The coated SS substrates were calcined at 450 °C to obtain the counter electrode (Scheme 1(b)).

2.4. Preparation of the solid electrolytes

For the solid-state polymer electrolyte preparation, PVC-*g*-POEM in THF was combined with MPII, LiI and I_2 in acetonitrile. A graft copolymer consisting of a PVC backbone and POEM side chains was synthesized through atomic transfer radical polymerization (ATRP) [37–39]. The salt mixture of LiI + MPII was fixed at a 1:1 wt ratio, and the ratio of the salt content was maintained at [EO]:[salt] = 10:1 [39].

2.5. Dye adsorption

Three types of indoline dye (Scheme 2. D102, D131, and D149; 0.5 M) in acetonitrile and butanol (1:1) were prepared for the ZnO NR-SS immersion [40]. The respective sensitizers were selectively adsorbed into ZnO NR-SS and then kept inside an oven at 65 °C with different loading times to determine the optimized immersion time for each dye.

2.6. Fabrication of the DSSTs

Scheme 3 shows the fabrication procedure for a single or multi-color DSST. A DSST with a satin weave structure was fabricated according to the method in a previous publication [36]. Two types of single or multi-color DSSTs were fabricated using a handloom. For each type of DSST, five threads of dye loaded ZnO NR-SS and five threads of Pt-SS were woven into a satin weave structure as the warp and weft, respectively. For the single-color-based DSST, all five threads of ZnO NR-SS were immersed in one dye color (D102, D131 or D149). Among the five ZnO NR-SS threads for the multi-color-based DSST, three counts were loaded with one main color, while the other two counts were loaded with the other two colors. Each single or multi-color-based DSST was named 102S, 131S, 149S, 102M, 131M or 149M according to the main color. A solid polymer electrolyte was carefully casted on the fabricated satin structure and finally coated with two layers of the PET film into a sandwich structure.

2.7. Characterization

Scanning electron microscopy (SEM, SUPRA 55VP, Carl Zeiss) was used to characterize the surface and cross sectional morphology of the ZnO NR arrays on an SS wire substrate. A UV–visible spectrophotometer (Hewlett–Packard, Hayward, CA, USA) was used to observe the absorption characteristics of each sensitizer. X-ray diffraction (XRD) measurements were performed

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