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Layered co-sensitization of gardenia and monascus for panchromatic light harvesting in dye-sensitized solar cells☆

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Abstract

TiO_2 electrodes adsorbed with two natural dyes (gardenia yellow and monascus) were used as sensitizers to improve the conversion efficiency of cocktail dye-sensitized solar cells (CDSC) for light harvesting over a wide range of wavelength. Adsorption and electrochemical properties of two dyes were evaluated based on adsorption kinetics and electrochemical measurements. In addition, the photovoltaic performance of a photo-electrode adsorbed with single-dye (gardenia yellow and monascus) or the mixture or successive adsorption of the two dyes, was evaluated from current–voltage measurements. Layered co-sensitization of the two natural dyes was compared depending on the adsorption modes. As for the TiO_2 electrode with successive adsorption of monascus and gardenia yellow dyes, the solar cell yields a short-circuit current density (J_{sc}) of 2.04 mA/cm^2, a photovoltage (V_{oc}) of 0.63 V, and a fill factor of 0.64, corresponding to an energy conversion efficiency (η) of 0.82%.
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1. Introduction

Dye-sensitized solar cell (DSSC) is comprised of an adsorbed dye, a nanoporous semiconductor electrode-adsorbed dye, an electrolyte, and a counter electrode [1–3]. The performance of DSSCs is mainly influenced by the dye as a sensitizer. Although many transition metal coordination compounds have been employed as sensitizers, they are expensive and environmentally toxic [4,5]. Recently, several natural organic dyes including chlorophyll, anthocyanin, tannin, and carotene extracted from various plants, fruits, flowers, and leaves have become a viable alternative to expensive and rare organic sensitizers [6–9]. Unfortunately, these natural dyes often work poorly in DSSCs. To overcome this problem, some researchers have employed two or more different dyes in such combination for absorption in lower and higher wavelength regions for the enhancement of DSSC performance [10–14]. Generally, a good dispersion of dye molecules dissolved in solvents could improve the efficiency of a DSSC. The solubility and adsorption capacity

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of natural dyes on an oxide surface are highly dependent on solvents. Therefore, it is meaningful to investigate the influence of solvents on the solubility of natural dyes for DSSC performance. Recently, our research group reported the influence of polar aprotic and protic solvents including water, ethanol, and dimethylsulfoxide (DMSO) on the solubility and photovoltaic conversion efficiency of monascus [15]. As a result, DMSO solvent showed a higher photovoltaic performance compared to other solvents of water and ethanol. Generally, unfavorable interactions between neighboring dye molecules on an oxide surface is observed in dye-cocktails, resulting in decreased photovoltaic performance [16,17]. It has been reported that the successive adsorption of dyes had a significant advantage over dye-cocktails in overcoming the problem of unfavorable dye interactions [18]. Therefore, it is meaningful to compare the simultaneous and successive adsorption of the dyes to enhance both the adsorption amount and the electrochemical properties.

To this end, the adsorption characteristics from electron harvesting point of view and electrochemical properties from electron transfer point of view using gardenia yellow dissolved in water and monoscus dissolved in DMSO, mixtures thereof, or successive adsorptions of both dyes were investigated. Adsorption kinetics data obtained under different adsorption modes were analyzed by employing a pseudo-second-order model. The electrochemical properties and photovoltaic performance of DSSCs using a single dye and mixtures of dyes were studied for harvesting light over a wide range of wavelengths.

2. Experimental

Gardenia yellow and monascus were purchased from the Naju Nature Dyeing Culture Center (Korea). The principal components of gardenia and monascus were reported in previous works [19,20]. Although the most active component of gardenia and monascus for adsorption and photoconversion in DSSCs have not identified in detail, the main components of gardenia are genipin, geniposide, geniposidic acid, rutin, ursolic acid, stigmasterol, and chlorogenic acid and those of monoscus are zanthomonascin_A1, zanthomonascin_A2, monascin, rubropunctatin, and rubropunctamine. Water and DMSO were used to dissolve the natural dyes of gardenia and monascus. The preparation of electrode, electrolyte and assembly of DSSCs was likewise described in our previous work [14]. A UV–vis spectrophotometer (Shimadzu UV-1601A, Japan) was used to inspect the absorption spectra of the natural dye solutions and the mixed solution. The adsorption kinetics experiments were carried out by immersing the TiO_2 cell with an active area of 4 cm^2 in a small adsorption chamber filled with 500 ppm aqueous solution of gardenia and monascus dyes. The experimental procedure was well described in our previous work [21]. The adsorption capacities of the TiO_2 films were determined by measuring the dye concentrations before and after adsorption using a UV spectrophotometer at 441 nm and 488 nm after filtration with a 0.015 m UF membrane filter. On the other hand, the current–voltage (*J–V*) curves of the fabricated DSSCs were measured using a source measure unit under irradiation of white light from a 200 W Xenon lamp (McScience, Korea). The incident light intensity and the active cell area were 100 mW/cm^2 and 0.25 cm^2, respectively. The *J–V* curves were used to calculate the short-circuit current density (J_{sc}), open circuit photovoltage (V_{oc}), fill factor (*FF*), and overall conversion efficiency (η_{eff}) of the DSSCs. Electrochemical impedance spectroscopy (EIS) measurements were performed using alternating current (AC) impedance (CHI 660A Electrochemical Work-station, USA) over the frequency range 1–10^5 Hz with amplitudes of ±5 mV over the V_{oc}.

3. Results and discussion

Fig. 1 shows photographic images and the UV–vis absorption spectra of the gardenia, monascus and the mixture of two dyes both dissolved in water and adsorbed on TiO_2 thin films. A clear difference in the absorption characteristics of gardenia and monascus dyes was observed. The absorption peaks for the gardenia extract was observed at a wavelength of approximately 441 nm, and those of cochineal at approximately 410 and 488 nm. A higher photovoltaic conversion efficiency in DSSCs adsorbed with both the gardenia and monascus dyes could be potentially achieved from the light absorption spectrum over a wide range of wavelengths.

Adsorption equilibrium and kinetics data provide valuable information for understanding the mechanism of the sorption process [19]. The adsorption kinetics of gardenia and monoscus dyes on the TiO_2 film from aqueous solutions were obtained and then analyzed by employing a pseudo-second-order model. Fig. 2(a) and (b) shows the obtained concentration decay curves of single and mixture (i.e., cocktail solution) adsorption of gardenia and monascus on the TiO_2 film obtained in a small adsorption chamber. The initial dye concentration and temperature were 500 mg/L and room temperature, respectively. The pH was not controlled. With increasing time, the amount of the adsorbed gardenia and monascus

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