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Original Research Article

Scattering Layers for Titanium Dioxide and Tin Dioxide Dye-sensitized Solar Cells with Pandan Leaf as a Natural Sensitizer

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ABSTRACT

Dye-sensitized solar cell with a scattering layer as a light mobility enhancer has been fabricated in this work. The cells were fabricated with two types of semiconductor materials which are Titanium dioxide and Tin dioxide. Natural sensitizer from Pandan leaves was also used as an electron generator combined with liquid electrolyte redox couple Iodide/Tri-Iodide. The photovoltaic performance was measured with four types of semiconductor configurations. It was Titanium dioxide only, Titanium dioxide with scattering layer, Tin dioxide only and Tin dioxide with scattering layer. Pandan dyes have higher photovoltaic performance when fabricated with scattering layers in both semiconductor cells. The scattering layer has boosted the light management inside the photo-anode film and increased the Photo-conversion efficiency. Photo-conversion efficiency parameters such as short-circuit current and open circuit voltage were monitored to improve compared to cells without the scattering layer. Higher electron injection were contributed from the increase of light path on photo-anode film with scattering layers. The incident photon-to-current efficiency graph confirmed that the cells with Titanium dioxide with scattering layer had a higher photon-to-current density than the cells with Tin dioxide only on 300 to 500 nm. The incident photon-to-current efficiency of the device with Tin dioxide with scattering layer was higher than that of the device with Tin dioxide only in the wavelength range of 300 to 700 nm, the dominant effect of the scattering layer was found to affect more in Tin dioxide than Titanium dioxide semiconductors on low to high wavelength of the incident photon-to-current efficiency. It concludes that light management is an important strategy to improve the performance of low-cost and eco-friendly solar cells. This strategy is suitable to enhance the efficiency of DSSC for further low-cost solar cells.

KEYWORDS

Dye-sensitized, Natural dye, Pandan leaf, Chlorophyll, Scattering layer, Light mobility.

INTRODUCTION

Dye-sensitized solar cells (DSSCs) offer a viable substitute to conventional photovoltaic devices due to their low-cost fabrication [1] conversion efficiency [2]. The basic component for a DSSC is a nanocrystalline semiconductor thin film dioxide coated with a light-absorbing dye [3]. Previous research reported various thin film of nanocrystalline semiconductor. In an effort to find alternatives to titanium dioxide (TiO_2), zinc oxide-tin dioxide (ZnO-SnO_2) based zinc stannate (Zn_2SnO_4) has emerged as a successful option. It has proven to be even better than ZnO and SnO_2 [4]. Researchers have explored an environmentally friendly aqueous solution synthesis route to fabricate small tin dioxide (SnO_2) nanoparticles (3-5 nm) at room temperature. This method does not require pressurized reaction vessels or organic solvents [5]. Another approach involves using zinc oxide (ZnO) nanostructure thin films, which have a large surface area, direct electron pathways, and effective light scattering centers. This technique can produce various ZnO nanostructures, including nanotubes, nanoporous structures, nanosheets, nanoflowers, nanoflakes, nanobranches, and nanolipsticks [6], [7]. In recent years, incorporating graphene sheets of different sizes in the photoanode has significantly improved the efficiency of DSSCs [8]. Additionally, carbon material [9], such as carbon counter electrodes for DSSCs, have attracted interest due to their low cost, excellent catalytic activity, and superior chemical stability. They are seen as potential alternatives to expensive platinum electrodes for large-scale DSSC applications. The dye absorbs photons and injects electrons into the conduction band of the semiconductor thin film oxides, creating an electric current. The electrons are then collected by an external circuit utilizing a catalyst layer [10] and returned to the dye through a redox mediator, such as iodide/triiodide, in a liquid electrolyte [11]. The DSSC has several advantages over conventional silicon-based solar cells, such as low cost, easy fabrication, and high efficiency under diffuse light conditions. The performance of a DSSC depends largely on the choice of the dye sensitizer, which should have a broad absorption spectrum in the visible range, a high molar extinction coefficient, good stability under light and heat, and a strong binding affinity to the semiconductor surface [12].

One of the topics that has attracted considerable attention in recent years is dye-sensitized solar cells (DSSCs) using natural dyes as sensitizers. DSSCs are a type of photovoltaic device that converts sunlight into electricity by using a dye molecule to absorb photons and inject electrons into a semiconductor. Natural dyes are derived from plants, fruits, vegetables, flowers, insects, etc., and have several advantages over synthetic dyes, such as low cost, environmental friendliness, abundance, and diversity [13].

The most widely used dyes in DSSCs are based on ruthenium complexes, which have excellent photophysical and electrochemical properties. However, ruthenium dyes are expensive and difficult to synthesize, which limits their large-scale application. Therefore, many researchers have been searching for alternative dyes, such as organic or inorganic dyes, that can act as co-sensitizers or replace ruthenium dyes altogether [14]. An aromatic electron acceptor has been employed to afford a near-infrared absorbing organic dye for a stable and efficient co-sensitized solar cell [15]. Other research has introduced six new cyclometalated ruthenium(II)-based dyes developed through ligand engineering to achieve higher power conversion efficiencies [16]. One of the advantages of organic dyes is that they can be easily modified by changing their functional groups or conjugation lengths to tune their optical and electronic properties. However, most organic dyes require anchoring groups, such as carboxylic acid or cyano-groups, to attach to the semiconductor surface via hydrophobic or covalent interactions [17]. Another source of potential dyes for DSSCs is natural dyes extracted from plants. Natural dyes have several benefits. The use of natural dyes in solar cells is a promising development to this technology because it cuts down the high cost of noble metals and chemical synthesis [18]. Therefore, this type of solar cell has attracted considerable attention from the academic and industrial communities. Various pigments, such as anthocyanin, carotenoid, chlorophyll, and flavonoid, derived from a variety of plant sources like leaves, fruits, and flowers, have been studied for their potential as sensitizers due to their easy availability [19]. Contrary to synthetic dyes, natural dyes are readily accessible, simple to prepare, cost-effective, non-toxic, environmentally friendly, and completely biodegradable. For example, various natural dyes can be combined to enhance the light absorbance spectra. Anthocyanin from purple cabbage and chlorophyll dye from spinach are extracted and mixed in a 1:1 ratio to create a sensitizer [20]. Another example was the successful synthesis of curcumin dye using potassium carbonate resulted in the production of a relatively stable deprotonated dye with a significant bathochromic shift in the visible light absorption spectrum, demonstrating potential for application in DSSCs [21]. Systems combining dyes and semiconductor materials with high biocompatibility are essential for improving efficiency [22]. Therefore, there is a need for more research on the development and optimization of natural dyes and DSSCs using natural dyes, as well as on the understanding of the underlying mechanisms and phenomena involved in the operation of these devices. This will help to overcome the existing challenges and limitations and enhance the potential applications of DSSCs using natural dyes in various fields, such as renewable energy, environmental protection, biotechnology, etc. In this study, the researchers examined the potential of pandan leaf extract as a dye sensitizer for DSSCs. Pandan leaves are commonly utilized in Asian culinary practices and are known for their distinctive green hue. The UV-visible absorption spectrum of the pandan dye solution in ethanol was measured, revealing a peak at approximately 670 nm, indicative of the chlorophyll content present in the leaves. Comparing with other chlorophyll source plant include spinach, alfalfa, parsley, wheatgrass, nettle, basil, and collard greens, the Pandan leaf has low economical value since the other chlorophyll source plants are food source. Pandan leaves are spiky and fan-shaped, and they thrive in tropical climates. The leaves are too fibrous and stringy to be consumed directly, but they possess an aroma similar to basmati rice and a subtle flavour similar to grassy vanilla and coconut. Pandan leaves are commonly pulverized to produce an emerald-green extract. The more mature the leaf, the darker the hue and deeper the flavour Pandan has been used in Asia for long time for colouring materials [23]. Additionally, a scattering layer was implemented as a means to enhance light mobility and optimize the absorption of light on the dye molecules.

EXPERIMENTAL METHOD

This section outlines the experimental methodologies employed in the present study. It begins with an overview of the dye material and its preparation, followed by a description of the fabrication and characterization processes involved in creating solar cells. The solar cells were manufactured and assessed within the Photofunctional Material and Device Laboratory at the Kyushu Institute of Technology in Japan.

Dye Material

Pandan is a plant species that belongs to the Pandanaceae family. It originates from Africa and has been introduced to the lowlands of Asia, Central and South America. Pandan leaves have various uses in Southeast Asia, such as herb tea and food colouring. Pandan also has medicinal properties and is used in traditional Ayurvedic medicine in India [23] and traditional Chinese medicine [24]. Pandan is a versatile and valuable plant that has many benefits for human health and well-being. The dye of pandan leaves is a green pigment called chlorophyll. Chlorophyll is a molecule that can absorb sunlight and convert it into chemical energy through the process of photosynthesis. Pandan leaves contain chlorophyll in special cells called chloroplasts. Chloroplasts are found in the mesophyll tissue of leaves, which is the layer between the upper and lower epidermis. The amount and type of chlorophyll in pandan leaves can vary depending on environmental factors, such as light intensity, temperature and humidity. Pandan leaves that grow in a bright and warm place usually have more chlorophyll than pandan leaves that grow in a dark and cold place. Chlorophyll gives pandan leaves their characteristic green colour, as well as their pleasant aroma and taste. Pandan leaves are often used as a natural colouring agent in cooking, cakes and drinks, because they can give a bright and attractive green colour.

Dye Preparation

The pandan leaf dye was prepared by washing the leaves with water and drying them in a shaded area for over three weeks. It then chopped the leaves into small pieces and ground them into a fine powder (20-180 mesh) using a Food Grinder (FTC-Z100). The next step, mix 10 grams of the powder with 100 ml of ethanol and sonicate the mixture for an hour. The mixture solution was stored in a dark place for three days and then filtered out the remaining powder using a vacuum pump. The dye solution kept in a dark glass container to avoid light exposure.

Preparation of dye-sensitized solar cell

Fluorine doped Tin Oxide (FTO) glass was used as a working electrode. Each glass was cut with dimensions of 15 × 20 mm and cleaned in an ultrasonic cleaner with distilled water, Isopropanol, and acetone for 15 minutes, sequentially. Titanium Dioxide (TiO_2) employed as a nanocrystalline semiconductor thin film coated on cleaned glass using a blade coating method. Working electrode variations were TiO_2 only and TiO_2 with a scattering layer (TiO_2+SL). For the TiO_2-only configuration, the cleaned FTO glass was coated with TiO_2 with a small particle size (Dyesol 30NRD) to facilitate dye adsorption. For the TiO_2 with scattering layer configuration, the cleaned FTO glass was coated sequentially with two types of TiO_2 paste: a large (400 nm) particle size paste to serve as a scattering layer and a small (30nm) particle size paste (Dyesol 30NRD) on top of it. Both configurations were sintered at 450 °C for 30 minutes after each coating and then immersed in pandan leaf dye solution for 24 hours for dye absorption.

The thickness of the working electrode TiO_2 layer was measured with SURFCOM130A with an average thickness of 7 µM for the TiO_2 only dan 10 µM for the TiO_2+SL configuration. The counter electrode was made of Platinum (Pt) sputtered on FTO glass. The working and counter electrodes were assembled in a sandwich structure with a 25 µm thick spacer (Solaronix Spacer) to create a gap for electrolyte injection. The electrolyte consisted of 500 mM of Lithium Iodide (LiI) and 50 mM of Iodine (I_2) as a base redox couple. 600 mM Ethylmethylimidazolium dicyanoimide (MeEtIm-DCA) was employed as an additive to enhance ionic conductivity [25] and 580 mM tert-butylpyridine (TBP) for open circuit voltage (Voc) and conversion efficiency enhancement [26]. The solvent used for the electrolyte was Acetonitrile.

The photovoltaic performance of the DSSCs was evaluated using a solar simulator (Bunko-Keiki Co. Ltd., Model Solar Simulator CEP-2000SRR). To prevent the light source from reflecting off the glass and reaching the cell from an angle other than the normal incidence, a black metal mask with a 0.214 cm^2 area was used in the photovoltaic measurement. This ensured that only the light that passed through the mask and hit the cell perpendicularly was measured, eliminating any optical effects that could distort the results. The mask had holes that matched the size and shape of the cell and was placed on top of the glass substrate.

RESULT AND DISCUSSION

This section provides an overview of the outcomes obtained from the experimental work conducted. It encompasses the analysis of dye material properties as well as the evaluation of solar cell performance. Furthermore, this section delves into a comprehensive discussion on a crucial approach aimed at enhancing the efficiency of affordable and environmentally sustainable solar cells.

Experimental Result

This study aimed to use Pandan leaf extract as a natural dye for sensitizing DSSC. The dye absorption spectra were measured by a UV-VIS spectrophotometer (JASCO V-670) and the results are presented in **Figure 1**. The peak wavelength of the dye was 665 nm, indicating a low solubility of Pandan leaf in ethanol. The dye extraction process took 72 hours to obtain a suitable concentration for DSSC fabrication.

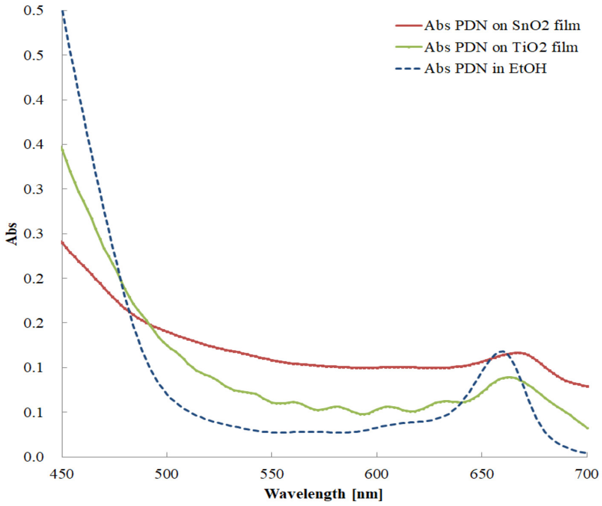

Figure 1. UV-Vis Absorption spectra of Natural dyes: Pandan in EtOH (dashed line) and on TiO_2 and SnO_2 films (solid line)

The performance of DSSC with TiO_2-only, SnO_2-only, TiO_2 with scattering layer (TiO_2+SL), and SnO_2 with Scattering layer (SnO_2+SL) were evaluated by the I-V curves shown in **Figure 2** and **Table 1**. The main factors affecting the Photoconversion efficiency (PCE) were the short circuit density (Jsc), the open circuit voltage (Voc), and the fill factors (FF). The PCE of the DSSC with TiO_2+SL was significantly higher than that of the DSSC with TiO_2 only, reaching 0.182% compared to 0.069%. This improvement was mainly due to the increase of Jsc from 0.203 mA/cm^2 to 0.481 mA/cm^2, which was attributed to the enhanced light harvesting and scattering effect of the additional layer [27]. Similar phenomena were observed in SnO_2-only and SnO_2 with a scattering layer (SnO_2+SL). The PCE of the DSSC with SnO_2+SL was significantly higher than that of the DSSC with SnO_2 only, reaching 0.143% compared to 0.052%. This improvement was mainly due to the increase of Jsc from 0.257 mA/cm^2 to 0.685 mA/cm^2. The Voc and FF values did not change much on both semiconductors TiO_2 and SnO_2 related to the addition of scattering layers.

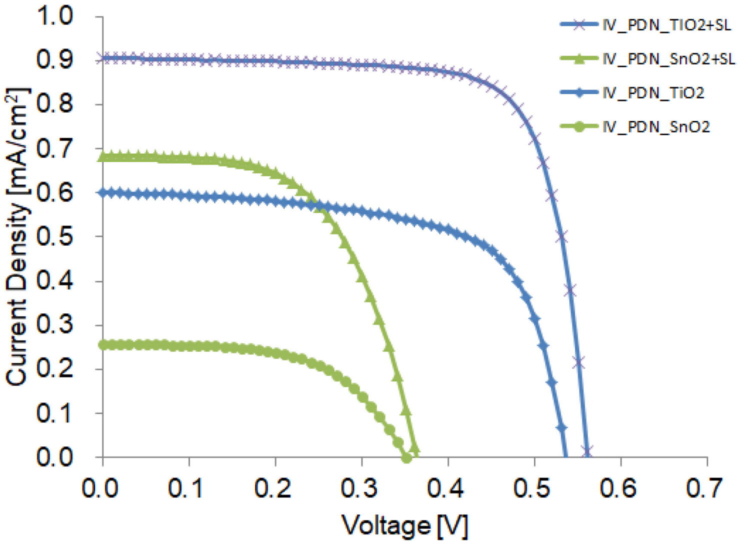

Figure 2. *I-V* Characteristic of DSSCs

As shown in **Table 1**, The enhanced performance of the device with TiO_2+SL and SnO_2+SL can be attributed to the higher photon-to-current conversion, as evidenced by the incident photon-to-current efficiency (IPCE) graph in **Figure 3**. The IPCE of the device with TiO_2+SL was higher than that of the device with TiO_2 only in the wavelength range of 300 to 500 nm, indicating more efficient electron injection from the SL to the TiO_2 layer. The IPCE of the device with SnO_2+SL was higher than that of the device with SnO_2 only in the wavelength range of 300 to 700 nm, the dominant effect of the scattering layer was found to affect more in SnO_2 than TiO_2 semiconductors on low to high wavelength of IPCE [26].

Table 1. I-V Characteristic of DSSCs with *Pandan* dye

	TiO_2	TIO_2 + SL	SnO_2	SnO_2 + SL
Efficiency [%]	0.212	0.382	0.052	0.143
FF	0.659	0.752	0.574	0.574
Voc [V]	0.535	0.560	0.350	0.363
Jsc [mA/cm^2]	0.601	0.906	0.257	0.685
Area [cm^2]	0.214	0.214	0.214	0.214

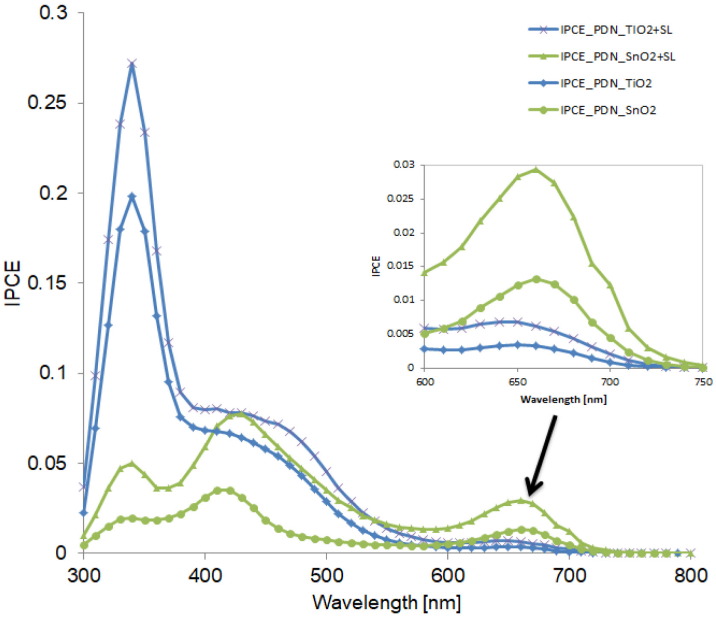

Figure 3. IPCE Spectra of DSSCs

Discussion

Dye-sensitized solar cells (DSSCs) using natural dyes as sensitizers still hampered in low efficiency. The light management is an important strategy to improve the performance of low-cost and eco-friendly DSSC. This strategy is proven to enhance the efficiency of device manufactured with scattering layer and without scattering layer as shown in **Table 1**. The optimalization of PCE for employment of scattering layer for TiO_2 and SnO_2 cells device was 80% and 175%, respectively. This mainly contributed by higher current induced in the system indicated by improvement on Jsc while Voc and FF relatively the same. This effect has been observed in previous works utilizing *Clitoria ternatea* flower as natural dyes with 163% PCE enhancements [28].

The effectiveness of this strategy has been confirmed not only for conventional TiO_2 semiconductors but also for shallower energy level SnO_2 semiconductors. When there is a mismatch between the Highest energy Occupied Molecular Orbital (HOMO) and Lowest energy Unoccupied Molecular Orbital (LUMO) of the dye molecule and the conduction band of the semiconductor used in dye-sensitized solar cells (DSSCs), it can result in a decrease in open-circuit voltage and overall photovoltaic performance. The energy levels of the HOMO and LUMO orbitals in the dye molecule play a crucial role in the functioning of DSSCs. To ensure efficient electron injection from the dye to the semiconductor in DSSCs, it is essential for the HOMO-LUMO energy levels of the dye molecule to align with those of the semiconductor. Any discrepancy between the HOMO-LUMO energy levels of the dye and the semiconductor can lead to suboptimal electron injection efficiency and diminished photovoltaic performance [29]. This discovery opens up new possibilities for the application of various natural dyes in DSSCs with different HOMO-LUMO band gaps.

CONCLUSION

The experiment on dye-sensitized solar cells (DSSC) using natural dye extracted from pandan leaf has been conducted. The experiment compared two types of semiconductors TiO_2 and SnO_2 and the addition of a scattering layer (SL). The results showed that the DSSC with TiO_2 and SL had a higher power conversion efficiency (PCE) than the DSSC with only TiO_2. The same phenomena are found in SnO_2 semiconductors. This was because the SL increased the light diffusion path on TiO_2 and SnO_2 which enhanced the chance of light interacting with the dye and injecting electrons into the semiconductor films. The optimalization of PCE for employment of scattering layer for TiO_2 and SnO_2 cells device was 80% and 175%, respectively. This mainly contributed by higher current induced in the system indicated by improvement on Jsc while Voc and FF relatively the same. The incident photon-to-current efficiency (IPCE) graph confirmed that the DSSC with TiO_2+SL had a higher photon-to-current density than the DSSC with only TiO_2 on 300 to 500 nm. IPCE of the device with SnO2+SL was higher than that of the device with SnO_2 only in the wavelength range of 300 to 700 nm, the dominant effect of the scattering layer was found to affect more in SnO_2 than TiO_2 semiconductors on low to high wavelength of IPCE. The effectiveness of this strategy has been confirmed not only for conventional TiO_2 semiconductors but also for shallower energy level SnO_2 semiconductors. This discovery opens up new possibilities for the application of various natural dyes in DSSCs with different HOMO-LUMO band gaps. It concludes that light management is an important strategy to improve the performance of low-cost and eco-friendly solar cells.

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