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Dye-sensitized solar cell using natural dyes extracted from spinach and ipomoea

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ABSTRACT

This study used spinach extract, ipomoea leaf extract and their mixed extracts as the natural dyes for a dye-sensitized solar cell (DSSC). Spinach and ipomoea leaves were first placed separately in ethanol and the chlorophyll of these two kinds of plants was extracted to serve as the natural dyes for using in DSSCs. In addition, the self-developed nanofluid synthesis system prepared a TiO_2 nanofluid with an average particle size of 50 nm. Electrophoresis deposition was performed to let the TiO_2 deposit nanoparticles on the indium tin oxide (ITO) conductive glass, forming a TiO_2 thin film with the thickness of 11.61 μm. This TiO_2 thin film underwent sintering at 450 °C to enhance the compactness of thin film. Finally, the sintered TiO_2 thin film was immersed in the natural dye solutions extracted from spinach and ipomoea leaves, completing the production of the anode of DSSC. This study then further inspected the fill factor, photoelectric conversion efficiency and incident photon current efficiency of the encapsulated DSSC. According to the experimental results of current–voltage curve, the photoelectric conversion efficiency of the DSSCs prepared by natural dyes from ipomoea leaf extract is 0.318% under extraction temperature of 50 °C and pH value of extraction fluid at 1.0. This paper also investigated the influence of the temperature in the extraction process of this kind of natural dye and the influence of pH value of the dye solution on the UV–VIS patterns absorption spectra of the prepared natural dye solutions, and the influence of these two factors on the photoelectric conversion efficiency of DSSC.

1. Introduction

The solar cells currently produced are generally divided into several types according to their material composition. Such as single-crystal silicon solar cells, multiple-crystal silicon solar cells, non-crystal-silicon solar cells and organic dye solar cells. The so-called “solar cells” generally refer to the cells made of silicon crystal material and their popularity is due to their higher photoelectric conversion efficiency compared to dye-sensitized solar cells (DSSCs). However, the production cost of the silicon-crystal-based solar cells are higher than the DSSCs. A dye-sensitized solar cell (DSSC) is a device to change light into electric energy by light sensitization established on wide energy-band semiconductor [1]. Simply due to their advantage of lower manufacturing cost, DSSCs are of great interest. With their simple and easy manufacturing process, DSSCs can be assembled in general environments at room temperature, and their cost is only around 1/3rd of the traditional silicon-based solar cells. In future, if the cost can be further reduced, when DSSCs are compared with silicon-based solar cells having the same efficiency, the total cost of DSSCs can be only 1/10th of that of silicon-based solar cells [2].

The efficiency of DSSC is determined mainly by the sensitizer used [3]. The marketed dyes for commercial use are mostly chemical synthetics, such as N719 and N3 dyes, both of which have satisfactory photoelectric conversion efficiency. However, these dyes always use some heavy metals, which are both expensive and produce environmental pollution. Generally, transition metal coordination compounds (ruthenium transition metal polypyridyl complexes) are used as effective sensitizers. This is because over the entire spectrum of visible light, there is intense charge-transfer absorption and metal to ligand charge transfer (MLCT) [4]. However, ruthenium polypyridyl complexes contain heavy metals, which make this kind of DSSC is unpopular from environmental aspects [5]. Moreover, the synthesis process of this complex is very complicated and expensive. In order to replace the rare and expensive ruthenium compounds, many kinds of natural dyes have been actively studied and tested as low-cost materials. However, natural dyes usually gave poor photovoltaic response in DSSC because of weak binding energy with TiO_2 thin film and low charge transfer absorption in the whole visible region range, but these dyes

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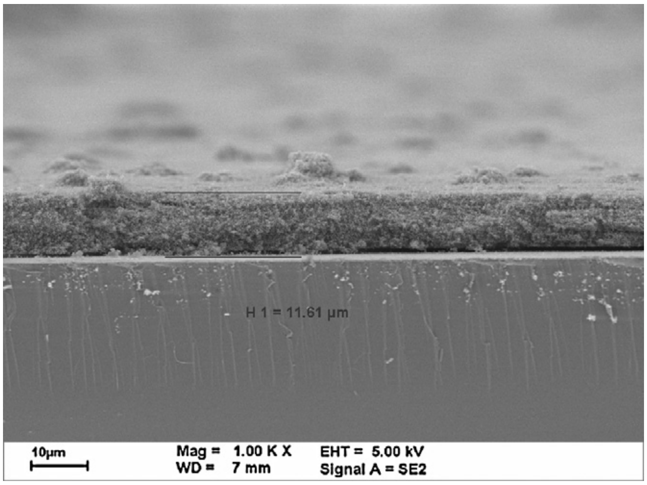

Fig. 1. FESEM image of the cross-section of the fabricated TiO_2 thin film.

are very cheap and can be prepared easily, compared to ruthenium polypyridyl complexes [6]. The energy conversion efficiency less than 1%, which is typical for most natural extracts tested so far. Several studies published in recent years have made a remarkable advance in the use of organic dyes for DSSCs [7,8]. For one, natural dyes can probably be applied to DSSCs, with the photoelectric conversion efficiency reaching a satisfactory level [9–11]. This is important since natural dyes have the advantages of high utility and very low cost.

In this research paper, extracts of ipomoea leaves and spinach were the natural dyes used as light sensitizers for the preparation of DSSCs. The leaves of most green plants are rich in chlorophyll [12], and the application of this kind of natural dye has been frequently investigated in many related studies. This paper stresses the effective extraction of dyes from these two plants and the preparation process of the dyes to enhance the overall photoelectric conversion efficiency of DSSCs.

2. Experimental procedures

2.1. Preparation of natural dye

First 10 g of fresh spinach and 10 g of fresh ipomoea leaves are separately put into 200 ml of alcohol each. Through indirect hydronic heating in boiling water, they were heated for 20 min to extract their chlorophyll. The solid dregs in the solution were filtered by filter paper to acquire a pure and natural dye solution. Then, the extract fluids of spinach and ipomoea leaves were blended at the ratio of 1:1 to serve as a natural dye mixture from these two plants.

2.2. Preparation of TiO_2 nanofluid

This research uses a novel processing technology to produce a nanocomposite fluid [13,14]. The arc discharge nanofluid synthesis system to produce a nanocomposite fluid was composed of a cooling system, arc discharge generator system, vacuum system, ultrasonic vibration system, nanofluid collector and pressure balancing system to fabricate a nanocomposite fluid, the anode and the cathode were pure Ti rod. The vacuum chamber was reduced to a vacuum level of around 30 Torr, and the processing started at the setting of breakdown voltage, peak current, pulse-on and pulse-off time. In addition, a nanofluid collector was attached with a vortex generator, enabling the nanoparticles to be evenly stirred by the vortex during collection. An Field Emission Scanning Electron Microscope (FESEM, LEO-1530) image was used to measure and analyze the structure and morphology of the prepared TiO_2 nanoparticles, and the XRD (X-ray Diffraction, MAC Science, MXP18) pattern was used to analyze the structure of the fabricated TiO_2 nanoparticles.

2.3. Preparation of DSSC photoelectrode

A TiO_2 nanofluid with particle size at 50 nm was prepared by the self-developed arc discharge nanofluid synthesis system. Then, the prepared TiO_2 fluid received electrophoresis deposition under normal temperature to deposit TiO_2 nanoparticles on the indium tin oxide (ITO) conductive glass, forming a TiO_2 thin film with thickness of 11.61 μm (as shown in Fig. 1). The active area of the DSSC is 0.25 cm^2 (0.5 cm × 0.5 cm). Since there were some electrolytes left on the surface of thin film, fissures were produced on the surface of the thin film. To improve this situation and increase the compactness of thin film, the TiO_2 thin film went sintering at 450 °C for 2 h. After sintering, the thickness of TiO_2 thin film is slightly decreased by 11.28 μm. Then the sintered TiO_2 thin film was immersed in the natural dye for 24 h, allowing the molecules of natural dye to be adsorbed on the surface of TiO_2 nanoparticles. Anhydrous alcohol was used to remove natural dye that had not been adsorbed on the surface of TiO_2 nanoparticles. Finally, after cleaning, the DSSCs photoelectrode was completed and ready for testing.

2.4. The assembly of DSSCs

Encapsulation was carried out according to the general assembly procedures (Fig. 2) of DSSCs. A DSSC is mainly comprised of ITO conductive glass, TiO_2 nanoparticles, natural dye, an electrolyte, counter electrode and spacers. For assembly, glass insulation spacers in long strips were first stuck on the four edges on the base plate of conductive glass at the bottom, forming a space between photoelectrode and counter electrode after assembly, and enabling the injection of electrolyte. Then AB glue was used to bind the base plate of lower conductive glass carrying the counter electrode with the base plate of upper conductive glass carrying the photoelectrode. The iodide electrolyte solution was injected from the edge of base plates. By capillary action, the electrolyte was absorbed between the photoelectrode and space until the whole space was filled. Then the injector inlet was sealed and the assembly of the DSSC was complete.

2.5. Measurement of photoelectric conversion efficiency of DSSC

A UV–VIS spectrophotometer (Jasco, V650) was used to inspect the absorption spectra of natural dye solution and the mixed solution of TiO_2 and natural dye. In addition, the photoelectric conversion efficiency of DSSC was inspected under the simulated sunlight source (AM1.5). With current–voltage (I–V) curve taken as the foundation, the fill factor (FF) was defined as follows:

$$\mathrm{FF} = \frac{I_{\max} \times V_{\max}}{I_{sc} \times V_{oc}} \tag{1}$$

where $I_{\max}$ and $V_{\max}$ denote the maximum output value of current and voltage respectively, and I_{sc} and V_{oc} denote the short-circuit current and open-circuit voltage respectively. The total energy conversion efficiency was defined as follows:

$$\eta = \frac{I_{sc} \times V_{oc} \times \mathrm{FF}}{P_{in}} \tag{2}$$

where P_{in} denotes the energy of incident photon.

3. Results and discussion

The process parameters were vacuum pressure of 30 Torr, peak current of 4 A, pulse-on and pulse-off times of 6 μs, and breakdown voltage of 220 V. Fig. 1 shows the FESEM image of the fabricated TiO_2 nanoparticle using the above-mentioned working parameters. As seen from the FESEM image in Fig. 3, the produced TiO_2 nanoparticle has a mean particle size of around 50 nm. By using the XRD pattern, the crystal structure of the fabricated TiO_2 nanoparticles could be seen. When producing the samples, a vacuum funnel was first used to filter out particles from the nanofluid. After desiccation, the acquired desiccated powder could act as the production sample. By comparing the XRD pattern (Fig. 4) after examination the standard spectrum of a JCPD card, the crystal structure of the fabricated particles was anatase TiO_2 (JCPD no. 21-1272).

Fig. 5 shows the acquired absorption spectra of ipomoea leaf extract, spinach extract and the mixed extract of these two plants after UV illumination. As seen from the curves of the spectra, the maximum absorption peak value of ipomoea extract fluid is at the wavelength of 410 nm, and the maximum peak value of spinach is at the wavelength of 437 nm. This figure clearly shows that the

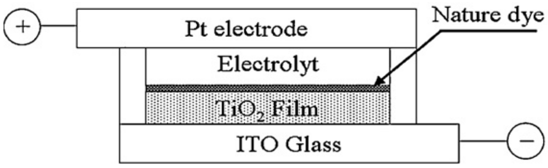

Fig. 2. Schematic diagram of the DSSC assembly.

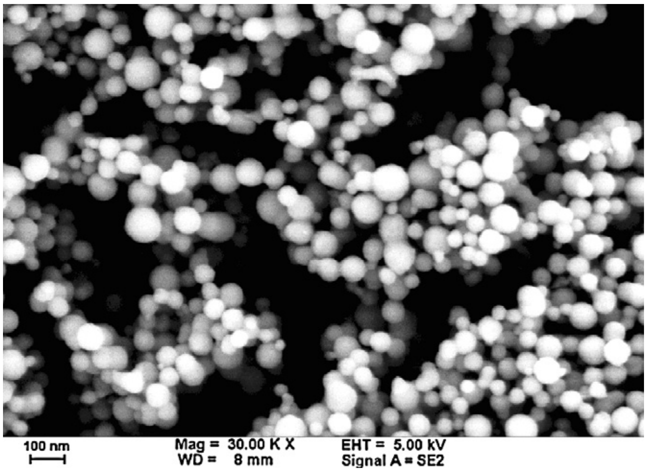

Fig. 3. FESEM image of the fabricated TiO_2 nanoparticles.

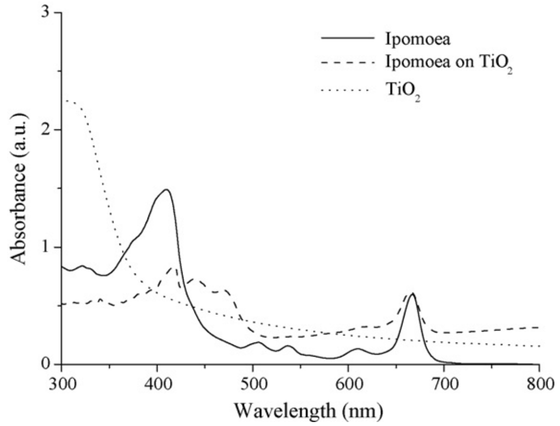

Fig. 6. Absorption spectra of ipomoea leaf extract fluid, ipomoea leaf extract fluid mixed with TiO_2 nanoparticles, and pure TiO_2 nanoparticles.

absorption properties of these two plants are obviously different at the wavelength range from 400 nm to 470 nm. Within this wavelength range, ipomoea leaves have the highest absorption peak, but their absorption range is narrower than that of spinach. The absorption spectrum of the mixed extract of these two plants is at the wavelength range from 400 nm to 500 nm, and the absorption intensity is obviously lower than that of spinach extract fluid. Furthermore, at the wavelength range from 500 nm to 700 nm, the difference of absorption spectrum intensity between the mixed extract, ipomoea leaf extract and spinach extract is not great. The spectrum property of the mixed extract is reduced mainly because of the different absorption properties of different pigments and the different colors of different extract fluids [15]. In addition, the difference in chemical structure of these pigments would also directly reflect the absorption spectrum.

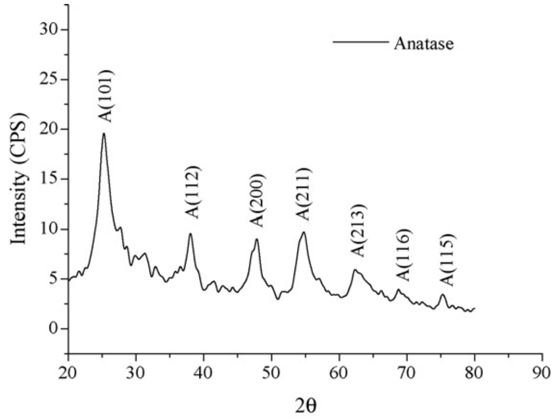

Fig. 4. XRD pattern of the fabricated TiO_2 nanoarticle.

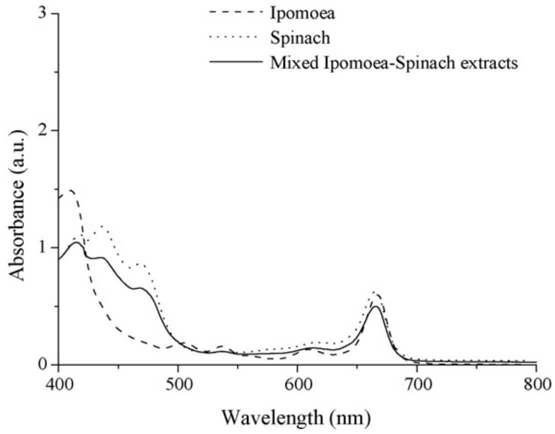

Fig. 5. Absorption spectra of ipomoea leaf extract, spinach extract and mixed extract of ipomoea leaves and spinach.

Fig. 6 compares the absorption spectrum of ipomoea leaf extract fluid with the ipomoea leaf extract fluid mixed with TiO_2 nanoparticles. It can be seen that after TiO_2 nanoparticles are added to ipomoea leaf extract fluid, its absorption intensity clearly decreases at the wavelength range from 300 nm to 440 nm, but clearly rises at the two wavelength ranges from 430 nm to 640 nm and from 690 nm to 800 nm. This property enables DSSCs to increase the charge-transfer ability under normal sunlight, giving them better absorption. Fig. 7 compares the absorption spectrum of spinach extract fluid with the spinach extract fluid mixed with TiO_2 nanoparticles. As observed from the figure, after TiO_2 nanoparticles are added to spinach extract fluid, its absorption intensity obviously decreases at the wavelength range from 300 nm to 480 nm, but obviously rises at the two wavelength ranges from 500 nm to 640 nm and from 690 nm to 800 nm. This is similar to the absorption property appeared in Fig. 6. Compare Fig. 6 with Fig. 7, the absorption peak value of the mixture of ipomoea leaves and TiO_2 is higher than that of the mixture of spinach and TiO_2 nanoparticles at the wavelength ranges from 300 nm to 500 nm. Fig. 8 shows the chemical structure of chlorophyll-a and chlorophyll-b contained in spinach and ipomoea leaves. Chlorophyll including chlorophyll-

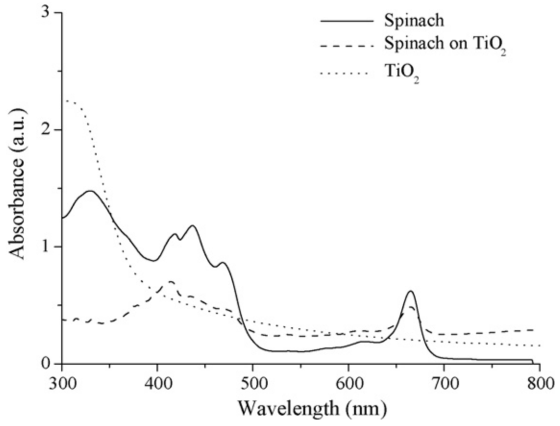

Fig. 7. Absorption spectra of spinach extract fluid, spinach extract fluid mixed with TiO_2 nanoparticles, and pure TiO_2 nanoparticles.

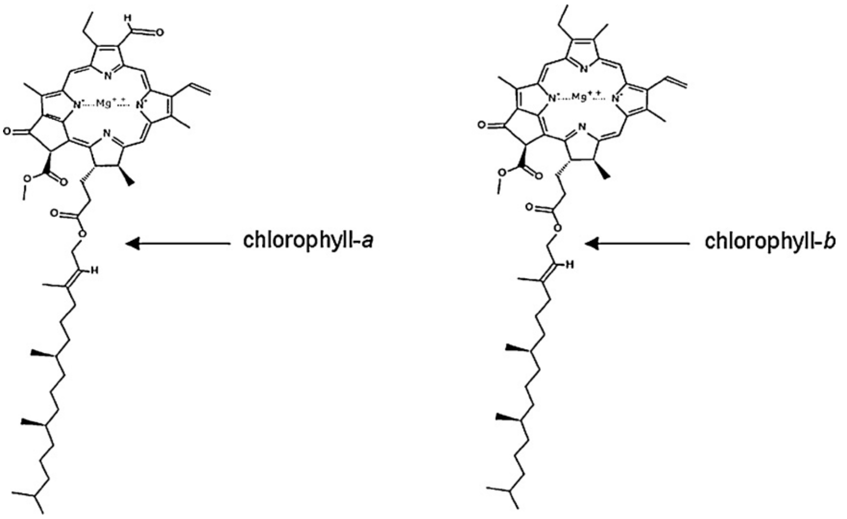

Fig. 8. Chemical structure of chlorophyll-a and chlorophyll-b contained in spinach and in ipomoea leaves.

Table 1
Photoelectrochemical parameters of the sensitized cells with natural extracts.

Extract	V_{oc} (mV)	J_{sc} (mA/cm^2)	η (%)	FF (%)
Spinach	550	0.467	0.131	51.00
Ipomoea	540	0.914	0.278	56.33

Table 2
Photoelectrochemical parameters of the sensitized cells with ipomoea leaf extract at various temperatures.

Extracting temperature (°C)	V_{oc} (mV)	J_{sc} (mA/cm^2)	η (%)	FF (%)
30	495	0.85	0.233	53.55
50	540	0.914	0.278	56.33
80	533	0.825	0.259	54.78

Table 3
Influence of pH of extract solutions on DSSC efficiency (under extraction temperature of 50 °C).

pH	V_{oc} (mV)	J_{sc} (mA/cm^2)	η (%)	FF (%)
3.0	510	0.915	0.253	55.15
2.0	543	0.982	0.292	56.38
1.0	565	1.12	0.318	59.23

a and chlorophyll-b is the main ingredient in the extract fluids of spinach and in ipomoea leaves. Therefore, the absorption spectrum property of the extract fluid is highly correlated to the chemical structure of chlorophyll [16–18]. Fig. 9 shows the *I*–*V* curve of the prepared DSSC that takes ipomoea leaf extract fluid as the natural dye. After calculation, its photoelectric conversion efficiency can reach 0.278%.

Furthermore, Table 1 compares different property parameters of DSSCs using different natural dyes, including short-circuit photocurrent density (J_{sc}), open-circuit voltage (V_{oc}), fill factor (FF) and energy conversion efficiency (η). As shown in Table 1, the photoelectric conversion efficiency of ipomoea leaf extract fluid is higher than the photoelectric conversion efficiency of spinach extract fluid. This is because, after the ipomoea leaf extract is adsorbed on the surface of TiO_2 nanoparticles, the absorption intensity is higher and the absorption wavelength range is broader than those after the spinach extract is adsorbed on the surface of TiO_2 nanoparticles. In addition, there is a higher interaction between TiO_2 nanoparticles and the chlorophyll in ipomoea leaf extract fluid, giving the produced DSSCs better charge-transfer performance, clearly improved efficiency. The reason is that the electrons can be transported from excited ipomoea leaf extract dyes molecule to TiO_2 thin film is much higher than the excited spinach extract dyes molecule to TiO_2 thin film [4]. Fig. 10 shows the incident photon-to-electron conversion efficiency (IPCE) for a DSSC sensitized with spinach and ipomoea leaf extract. As known from Fig. 10, when the wavelength of incident light is at the range of 300 nm to 400 nm, ipomoea leaf has higher absorption from the energy of incident light. Therefore, ipomoea leaf has higher conversion efficiency towards the energy of

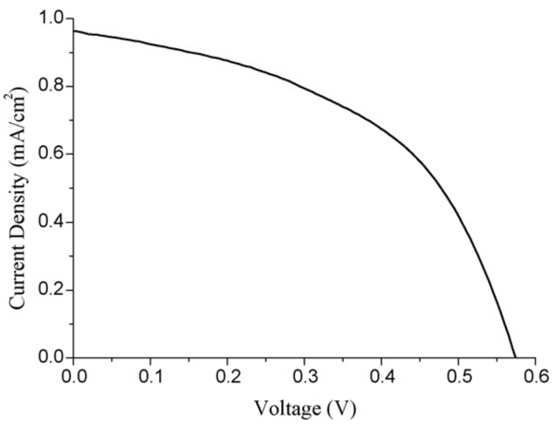

Fig. 9. Current–voltage curve for a solar cell sensitized with ipomoea extract.

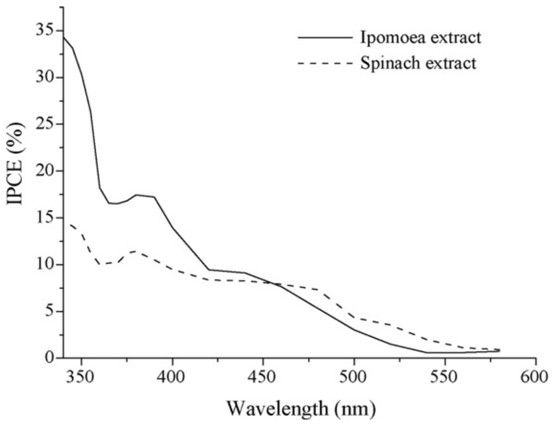

Fig. 10. IPCE curves for DSSCs sensitized with spinach and ipomoea leaf extracts.

Table 4
Photoelectrochemical properties of solar cells with natural extracts.

Dye	V_{oc} (mV)	J_{sc} (mA/cm^2)	P_{max} (mW/cm^2)	FF (%)	Refs.
Spinach	550	0.46	0.25	51	–
Ipomoea	540	0.91	0.36	56.33	–
Skin of Jaboticaba	660	2.6	1.1	62	[16]
Black rice	551	1.14	0.372	52	[4]
Sicilian orange	340	3.84	0.68	50	[23]
Skin of eggplant	350	3.40	0.44	40	[23]

incident light, and the short-circuit current density (J_{sc}) excited by ipomoea leaf is higher than spinach. Under the same preparation procedures and the same light illumination condition (AM 1.5), with ipomoea leaf extract fluid selected as the dye, and the extraction temperatures set at 30 °C, 50 °C and 80 °C, this study compared the influence of extraction temperature on DSSCs [19]. From Table 2, it can be seen that when the extraction temperature is 50 °C, the DSSCs with ipomoea leaves as the dye have the highest photoelectric conversion efficiency of 0.278%. This is because the chlorophyll-a and chlorophyll-b in the extraction fluid has the best stability and the degradation rate of pigment (chlorophyll-a and chlorophyll-b) is the slowest under the extraction temperature at 50 °C [20]. The table shows that the extraction temperature influences the charge-transfer efficiency of natural dye, implying that it also influences the sensitization effect, thus having direct influence on the photoelectric conversion efficiency of DSSCs. Therefore, in order to acquire the best photoelectric conversion efficiency, the extraction temperature of natural dye has also must to be controlled.

Furthermore, the paper investigated the influence of the pH value of extraction fluid on DSSCs, and compared the influences among the ipomoea leaf extract fluids with different pH values (see Table 3). Table 3 shows that when the pH value falls, the performance of DSSCs rises clearly; and when the pH value falls from 3.0 to 1.0, the photoelectric conversion efficiency of DSSCs is enhanced from 0.253% to 0.318%. Although absorption spectra of the extract fluid have the phenomenon of blue shift under acidic environment, it can enhance the fill factor of DSSC (as shown in Table 3) and the stability of natural dye molecules [21,22]. Therefore, the photoelectric conversion efficiency of DSSCs can be raised.

Table 4 shows the comparison of major parameters for the application of different natural dyes to DSSC in the past literature [4,23]. As seen from the table, the two natural dyes adopted in this study have lower J_{sc} than other natural dyes, but have higher values of V_{oc} and FF. Besides, the DSSC prepared by the study is made by the way of simple packaging. Under irradiation at various times, the change of short-circuit current density for the prepared DSSC is measured. The experimental result shows that after irradiation for 30 min, J_{sc} falls from the original 0.91 mA/cm^2 to 0.73 mA/cm^2; and after irradiation for 120 min, J_{sc} falls to 0.45 mA/cm^2. From these results, it is known that after the DSSC using natural dye has been under irradiation for a long time, there is still needed extremely great improvement for maintaining the stability of its photoelectric conversion efficiency.

4. Conclusions

Following the above experimental results and related discussion, the following conclusions can be drawn:

1. The TiO_2 nanoparticles prepared by the self-developed arc discharge nanofluid synthesis system had a particle size of 50 nm and good size consistency. Furthermore, the electrophoresis deposition technique was used to deposit the prepared TiO_2 nanoparticles on the indium tin oxide (ITO) conductive glass, forming a TiO_2 thin film with thickness of 11.61 μm.
2. The photoelectric conversion efficiency of ipomoea leaf extract fluid as the natural dye for the prepared DSSC can reach 0.278%. In addition, the ipomoea leaf dye extracted under extraction temperature of 50 °C and pH value of extraction fluid at 1.0, could achieve a photoelectric conversion efficiency of 0.318%.
3. As known from the IPCE curve, within the wavelength of incident light is at range 300–400 nm, since ipomoea leaf extract fluid has higher conversion efficiency towards the energy of incident light, the short-circuit current density excited by ipomoea leaf is higher than spinach, making the photoelectric conversion efficiency of ipomoea leaf become higher than spinach.

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