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Natural Dye Sensitizer in Dye Sensitized Solar Cell

(Pemeka Pewarna Semula Jadi dalam Pekaan Pewarna Sel Suria)

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ABSTRACT

Blue-pea flower, turmeric, mulberries, brown rice, purple cabbage and Indian mulberry leave were successfully form on TiO_2 mesoporous film using immersion method to produce TiO_2 mesoporous photoanode for natural dye sensitized solar cells (DSSCs) assembly. The TiO_2 mesoporous films were formed after calcinations at 450°C for 30 min. The photoanodes were dipped in different types of natural dye for 24, 72 and 120 h. The properties of natural dye were investigated by ultraviolet-visible spectroscopy (UV-vis) and Fourier transform infrared spectroscopy (FTIR). From UV-Vis spectroscopy analysis, the wavelength range of the natural dye studied in this research lays between 350 and 800 nm. The FTIR result of the natural dye shows the present of intermolecular H-bond, C=O stretching vibration, C-O-C stretching vibration, C=C bending and C-H bending which was due to the component of anthocyanin, carotenoids and chlorophyll. The characterization including field emission scanning electron microscopy (FESEM), energy dispersive x-ray (EDX) and x-ray diffraction (XRD) were carried out on the TiO_2 mesoporous film. On the other hand, the conductivity of electrolyte for liquid electrolyte, gel electrolyte and solid electrolyte were also investigated. Gel electrolyte has the highest conductivity, 26.1 mS/cm while liquid electrolyte and solid electrolyte obtained 17.34 and 0.45 mS/cm, respectively. Finally, solar cells were prepared by sandwiching the TiO_2 mesoporous photoanode with Platinum (Pt) counter electrode. The results showed short circuit current, open circuit current voltage, fill factor and efficiency for all samples during the present of light. The highest efficiency was obtained from Blue-pea sample that immersed for 120 h with 0.123% efficiency.

Keywords: DSSCs; natural dye sensitized solar cells; solar energy

ABSTRAK

Bunga telang, kunyit, malberi, beras perang, kubis ungu dan daun mengkudu telah melekat pada lapisan TiO_2 berliang meso dengan menggunakan kaedah rendaman bagi menghasilkan TiO_2 berliang meso sebagai fotoanod untuk pewarna semula jadi sebagai pemeka dalam pewarna sel solar berkepekaan (DSSCs). Lapisan TiO_2 berliang meso terbentuk selepas pengkalsinan pada suhu 450°C selama 30 min. Fotoanod dicelup dalam pewarna semula jadi selama 24, 72 dan 120 jam. Sifat pewarna semula jadi telah dicirikan oleh Ultra Violet Visible Spektrofotometer (UV-vis) dan spektroskopi Fourier infra merah (FTIR). Melalui UV-Vis spektroskopi, gelombang pewarna semula jadi yang diperoleh adalah antara 350 dan 800 nm. Hasil FTIR pewarna semula jadi menunjukkan terdapat ikatan H, C=O, C-O-C, C=C dan C-H yang disebabkan oleh komponen daripada pada antosianin, karotenoid dan klorofil. Medan pancaran mikroskop elektron imbasan (FESEM), tenaga serakan sinar-x (EDX) dan pembelauan sinar-x (XRD) telah dijalankan ke atas lapisan TiO_2 berliang meso. Konduktiviti elektrolit cecair, elektrolit gel dan elektrolit pepejal juga telah dikaji. Elektrolit gel mencatat konduktiviti tertinggi, 26.1 mS/cm manakala elektrolit cecair dan elektrolit pepejal memperoleh 17.34 dan 0.45 mS/cm. Fotoanod TiO_2 berliang meso dicantum dengan elektrod kaunter Platinum (Pt). Nilai yang ditunjukkan adalah semasa litar pintas, voltan litar terbuka, faktor pengisian dan peratus keberkesanan untuk semua sampel. Bunga telang yang telah direndam selama 120 jam telah mencatat peratus keberkesanan yang tertinggi iaitu 0.123%.

Kata kunci: DSSCs; pewarna semula jadi sel solar berkepekaan; tenaga solar

INTRODUCTION

The history of the sensitization of semiconductors to light of wavelength longer than that corresponding to the band gap has been presented elsewhere (Kim et al. 2011). Dye-sensitized solar cells or DSSCs use a process similar to photosynthesis to produce electrical energy. Dye-sensitized solar cells are a promising technology because they are inexpensive and resilient, making them ideal for large scale and small scale applications. However, they have lower efficiencies than most other types of solar cells, so they require more space than other types of solar cells to produce the same amount of electric energy. Today, most solar cells manufactured in the world are silicon solar cells. There are several types of silicon solar cells, one of which are bulk silicon solar cells and thin-film solar cells. One of the next-generation solar cells expected to satisfy these demands is dye-sensitized solar cells. Grätzel (2013) reported high-efficiency cells using nanoporous titanium oxide semiconductor electrodes, ruthenium (Ru) metal complex dyes and iodine electrolyte solutions in the journal of Nature in 1991. Since then, many studies have been actively carried out on DSSCs and showed their performance comparable to amorphous silicon thin films. These DSSCs have the advantages of low cost, lightweight and easy fabrication, but issues include durability and further improvement of their properties. In order to respond to these issues, many attempts have been made, such as solidifying electrolytes and improving materials and structures, but there have been no great breakthroughs yet. Dye-sensitized solar cells consist of dye-adsorbed porous TiO_2 layer on fluorine doped tin oxide (FTO) glass as working electrode. Under the illumination of solar light, dye molecules are excited and electrons were produced. In order to generate meaningful electrical power from DSSCs, the electrons need to pass four important interfaces of DSSCs: dye/ TiO_2, TiO_2/FTO, electrolyte/counter electrode and dye/electrolyte. The nanoporous nature of the TiO_2 layer provides high surface area that is great importance to the efficient photon-to-electricity conversion because it enhances dye loading and solar light absorption (Indriana Kartini et al. 2001). However, it also provide abundant TiO_2 surface site (direct route) and bare FTO conducting site (indirect route), where the photo-injected electrons may recombine with electrolyte. The recombination will cause the loss of the photocurrent. So the photovoltaic performance of DSSCs was seriously decreased (Cameron & Peter 2003). The introduction of a compact layer between the interfaces of the FTO/porous TiO_2 has been proven theoretically and practically effective to block the electrons recombination via the indirect route as shown in Figure 1 (Haque et al. 2004; Nakade et al. 2004, Zhu et al. 2006). Efficiencies over 10% have been achieved (Grätzel 2013). Therefore, the natural dye as sensitizer in dye sensitized solar cell is introduced. Metal-free dyes such as derivatives (Li et al. 2006), coumarin dye (Hara et al. 2001), porphyrin dyes (Tokuhisa & Hammond 2003) and cyanine and merocyanine dye (Kazuhiro Sayamaa, 2003) have been used as sensitizers, but did not achieve the same solar conversion efficiency as ruthenium dyes. In this research, blue-pea flower, turmeric, mulberries, brown rice, purple cabbage and Indian mulberry leave were used and successfully form on TiO_2 mesoporous film using immersion method in order to produce photoanode for natural dye sensitized solar cells (DSSCs). The highest efficiency achieved in this work was 0.123% which was higher than reported value for ruthenium dyes. In addition this dye has the advantage which can be produced at low cost, as it does not contain the rare metal ruthenium and easy to synthesize.

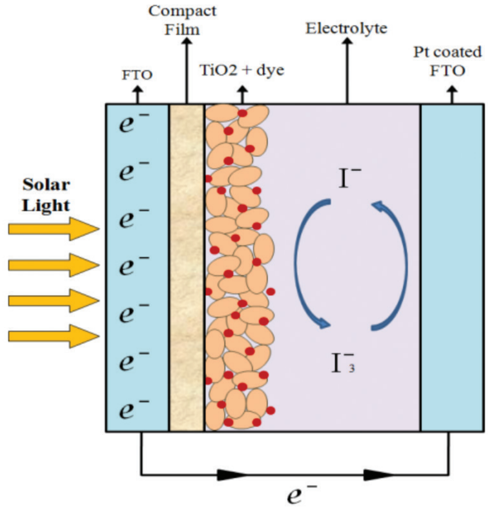

FIGURE 1. Schematic diagram of dye sensitized solar cells

EXPERIMENTAL DETAILS

PHOTOANODE PREPARATION

In this research, fluorine tin oxide (FTO) with sheet resistance ~7 Ω/square was used as substrates. The FTO substrates were cleaned with distilled water and soap (Extran) and then rinse with distilled water and sonicated for 15 min. The TiO_2 mesoporous photoanodes were selected as the working electrode due to exhibited best operating performance in classical DSSCs. In this research the TiO_2 mesoporous layer was deposited by doctor blade using commercialised Dyesol paste 18NR-T. The substrates were then sintered at 450°C in the Lenton furnace in order to achieve good contact between FTO and TiO_2 nanoparticles.

PREPARATION OF NATURAL DYE SENSITIZERS

The blue-pea flower, turmeric, mulberries, purple cabbage and Indian mulberry leave were cut into small pieces. Each natural dye sources were weight for 50 g and soak with 124 mL of ethanol and boil for 30 min. The beaker should be cover with aluminium foil or plastic sheet in order to prevent ethanol from evaporate. Then, the solutions were cool to room temperature for 24 h. After that, by using filter funnel and conical flask, the solution was filtered in order to remove solid fragments. The extract solution of all natural dyes (Figure 2) will appear, for blue-pea flower it has absolutely blue colour of solution. The solution was then kept in the amber bottle to avoid the solution from direct sunlight. After the filtration process, the FTO substrates with TiO_2 transparent layer were dipped in natural dye solution for 24, 72 and 120 h. However, for brown rice, there was no need to boil the mixture of brown rice with ethanol. First, the brown rice was weight for 50 g. Then, it was cleaned with distilled water. Let it dry in room temperature for one day. Next, soak together the 50 g brown rice with 124 mL of ethanol in beaker for 48 h with the ratio of 1:1. After that, filter the solution using the filter funnel and conical flask. The filter paper used is Grade 4 which is for rapid filtering with excellent retention of coarse particles. The filtration process was done in the fume cupboard. After the filtration process, FTO glasses that deposited with TiO_2 were dipped in the solution for 24, 72 and 120 h. The beaker must be properly stored and protected from direct sunlight.

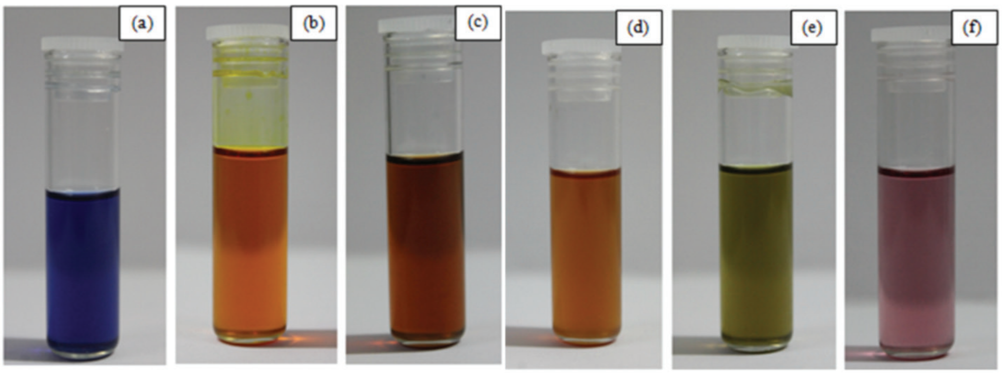

FIGURE 2. Extracted dyes of natural dyes such as (a) blue-pea flower, (b) turmeric, (c) mulberries, (d) brown rice, (e) Indian mulberry leaves and (f) purple cabbage

COUNTER ELECTRODE

Platinum was used as a counter electrode. Platinum is known for its strong catalytic activity in electron transfer at the counter-electrode. An efficient cathode for a Dye Solar Cell can consist of a thin platinum layer on a FTO substrate. By using the doctor-blading technique, the platinum was coated on the FTO substrates and heated up at 450°C on a hot plate before using them. This step was to activate the catalytic platinum layer. The working electrode should be used right after preparation. Put the electrodes against each other so that the stained TiO_2 faced the platinum of the counter electrode. The conductive sides of each electrode, face to face, form the inside of the cell. Use paper binders or similar clips to hold the electrodes together. The cell should be immediately filled with electrolyte before getting damaged by ambient air.

RESULTS AND DISCUSSION

CHARACTERIZATION OF FTO SUBSTRATE AND TIO_2 MESOPOROUS FILM

Field emission scanning electron microscope (FESEM) was used to study the surface morphology of the sample. However, FESEM image could hardly provide reliable information as it was difficult to conclude that the images observed were referred to the FTO substrate or TiO_2 mesoporous film. Therefore, the FESEM was equipped with Energy Dispersive X-ray Spectroscopy (EDX) analysis to determine the presence of different element in the samples. Based on these observations, the surface roughness were depends markedly on the fabrication parameters. The variation in the transmission can be explained mainly by a variation in light scattering due to surface roughness. Figure 3(a) presents the view of the surface of FTO substrates that was used. Under the optimum conditions, it can be seen that FTO substrates were adequate as highly conductive and transparent layers. For the technology of natural dye in solar cell, FTO has been chosen for its better properties and its stability during the processes (R. Pommier 1981). The EDX determined the presence of different element in the sample by providing relative proportions of the element in weight or atomic percentage on the surface or subsurface either in spot or square scan area analysis. The EDX spectra were performed at 10KX and 30KX magnification for the samples. By referring to the analysis of EDX for FTO substrate in Figure 3(b), Sn has the highest wt. % of 70.57, followed by 16.05 wt. % of oxygen, 13.10 wt. % of titanium and lastly, 0.28 wt. % of chlorine. Based on EDX results, there was no weight percent of fluorine because the amount was too little. It was important to have a smooth FTO substrate without pores and cracks to ensure high performances of TiO_2mesoporous film. The EDX detected the presence of element associated with titanium (Ti), oxygen (O) and tin (Sn) in the TiO_2 mesoporous film. This further confirmed the presence of both TiO_2 and FTO substrate, which was also detected in the XRD result. There were 61.45 wt. % titanium (Ti), 34.84 wt. % oxygen (O) and 3.71 wt. % tin (Sn) for TiO_2 mesoporous film (Figure 3(c)). Supposedly, there should be 40 wt. % of O, but it was only 34.84 wt. % because EDX gives qualitative analysis. Basically, the presence of Sn element is from the FTO substrate. Based on the result, Sn element was from the FTO substrate while Ti element was from the TiO_2 paste. The X-ray diffraction (XRD) patterns shows that, at 450°C, film were two different phases of TiO_2 were observed, both brookite and anatase, in varying ratios across the substrate. An example strip of the X-ray patterns, showing the presence of both phases, is depicted in Figure 4. The anatase TiO_2 peak were detected at 2θ of 25.20˚, 33.46˚, 37.44˚, 48.01˚, 54.07˚, 61.25˚ and 70.40˚ corresponding to diffraction plane of (101), (103), (004), (200), (105), (213) and (220), respectively. However, FTO peak were detected at 2θ of 26.23˚, 51.27˚, 65.32˚, 78.00˚ and 80.40˚ corresponding to diffraction plane of (111), (240), (213), (431) and (124). Consideration of all of the diffraction patterns found that anatase was the dominant phase; present to some extent in all of the spots and only small amounts of brookite detectable.

CHARACTERIZATION OF NATURAL DYE

Ultraviolet-visible spectroscopy (UV-vis) The wavelength range of the absorption spectrum lay between 350 and 800 nm. The related spectrum was shown in Figure 5.

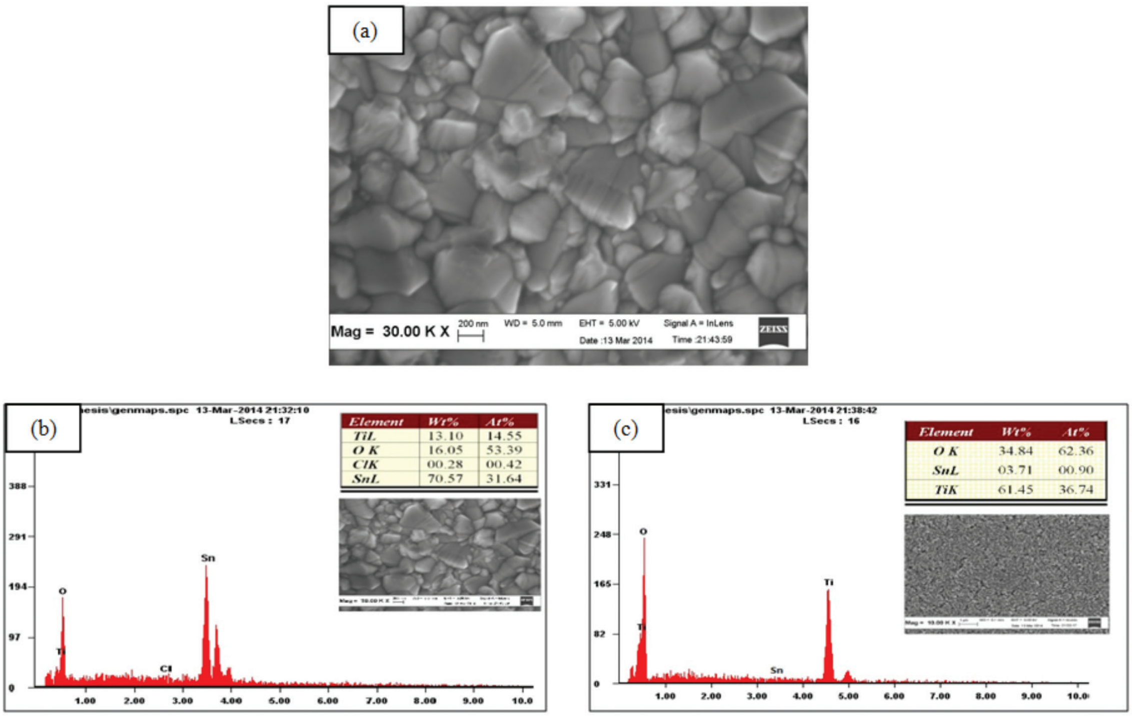

FIGURE 3. (a) Scanning electron micrographs of FTO layer, (b) EDX analysis of FTO substrate, and (c) EDX analysis of TiO_2 mesoporous film

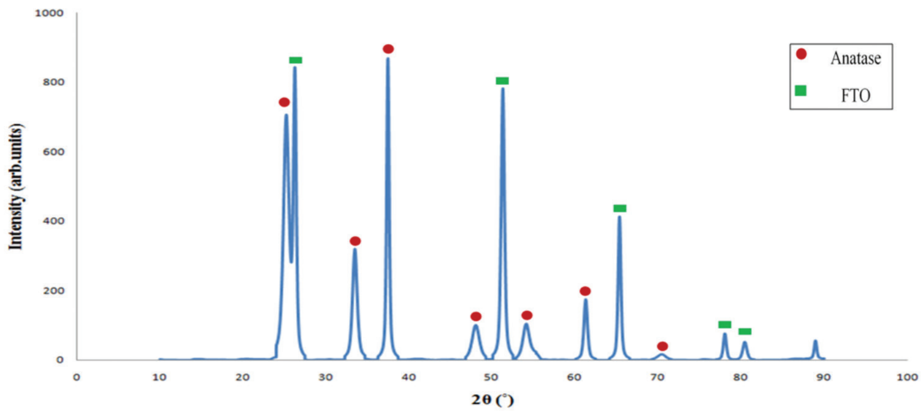

FIGURE 4. X-ray diffraction patterns of TiO_2 mesoporous film

The wavelength and absorbance for each of natural dyes were compared to commercial dye which is N719. The light absorbance was most probably due to the colour pigments of the natural dye itself. More important than their reflection of light is the ability of pigments to absorb certain wavelengths.

The dye have different wavelength due to different molecular structure; anthocynin, carotenoids and chlorophyll. Basically, this absorption is due to the anthocynin, carotenoids and chlorophyll obtained from natural dyes. In Table 1, the wavelength of natural dyes can be compared to the wavelength of commercial dye which is N719. As shown in Table 2, the peak of absorbance can be found at three wavelengths which were at 310, 382 and 520 nm. N719 was use as reference because of its excellent performance in terms of efficiency and stability. Dyesol's N719 industry standard dye is a modification of the N3 foundation dye with changes that increase cell voltage. Like all Dyesol dyes, Dyesol's N719 industry standard dye offers guaranteed performance, high reproducibility, stable/consistent results and is of the highest purity. Therefore, the natural dyes that contain anthocyanins molecular structure such as, mulberries, blue-pea flower, brown rice and purple cabbage tend to have high performance due to their wavelength that almost the same with N719 which was around 500 to 600 nm.

Fourier Transform Infrared Spectroscopy (FTIR) The result from FTIR in Figure 6 shows a few of peak in absorbance wavenumbers spectrum range from 4000 to 550 cm^{-1}. From

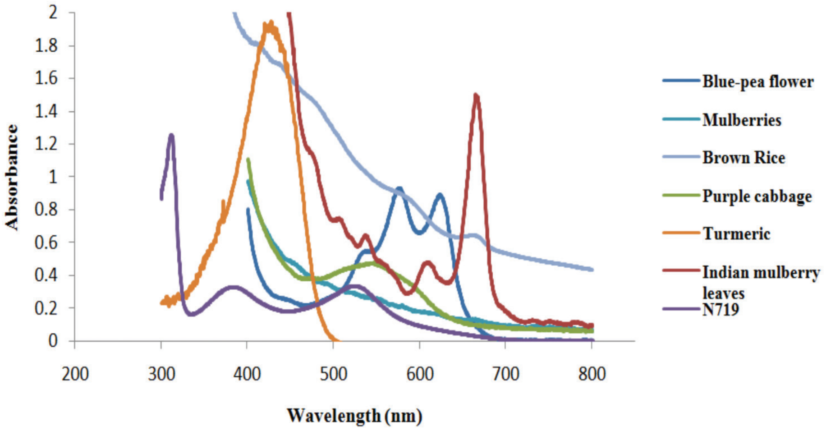

FIGURE 5. The absorption spectra of the extracts of blue-pea flower, Indian mulberry leaves, purple cabbage, mulberries, turmeric, brown rice and in ethanol solution

TABLE 1. Natural dye, wavelength and pigments

Natural dye	λmax (nm)	Molecular structure
Blue-pea flower	574	Anthocyanin
Turmeric	427	Carotenoids
Mulberries	580	Anthocyanins
Brown rice	553	Anthocyanin
Purple cabbage	542	Anthocyanin
Indian mulberry leaves	665	Chlorophyll

TABLE 2. Wavelength of N719

Dye	Wavelength, λmax (nm)	Description
N719	λ1 = 310 λ2 = 382 λ3 = 520	• Modified dye to increase device voltage • High performance dye

the result obtained, all samples have broad absorption range between 3200 and 3400 cm^{-1} showed the chemical has intermolecular hydrogen bond (H-bond). Almost all the samples have sharp absorption between 1600 and 1700 cm^{-1} show that C=O stretching vibration is conjugate, except turmeric. The sharp peak at 1030 ~ 1060 cm^{-1} for all samples showed that there was C-O-C stretching vibration of ester acetates. Based on the result, it was showed that the pattern of FTIR result for blue-pea flower, mulberries, brown rice and purple cabbage were almost the same. These results proved that blue-pea flower, mulberries, brown rice and purple cabbage contained the anthocyanin which is the core composition for natural dye in DSSC. The carbonyl and hydroxyl group in blue-pea flower, mulberries, brown rice and purple cabbage can be bound with the surface of TiO_2 film and thus result in photoelectric conversion effect (Riyaz & Nafarizal 2010). There were some differences for turmeric and Indian mulberry leaves since both of these plants contain difference molecular structure of pigments, carotenoids and chlorophyll. The sharp absorption of FTIR result for turmeric at 1550 ~ 1580 cm^{-1} showed that C=C bending. Absorbance range between 860 and 680 cm^{-1} in turmeric show that there was aromatic C-H bending.

CHARACTERIZATION OF CELL

Current Density-Voltage (J-V) Measurement Current density-Voltage (J-V) measurement was carried out using Keithley source meter model 4200 under radiation of 150W Xenon lamp. Then, fill factor (FF) and efficiency (η) of the cells were calculated based on J-V curve. Light intensity used in this research was 1000 W/m^2. Table 3 shows the efficiency of all samples based on the immersion time.

Compared to all the samples that contained anthocyanin molecular structure, blue-pea has the highest performance followed by mulberries, brown rice and purple cabbage. As can be seen from Table 3, all the samples for every naturals dye with the immersion time of 120 h gave the maximum photo responses in dye sensitized solar cell. The reason for this was because the formation of multiple layers of dye molecule or the aggregation of dye molecules around the TiO_2 crystallites.

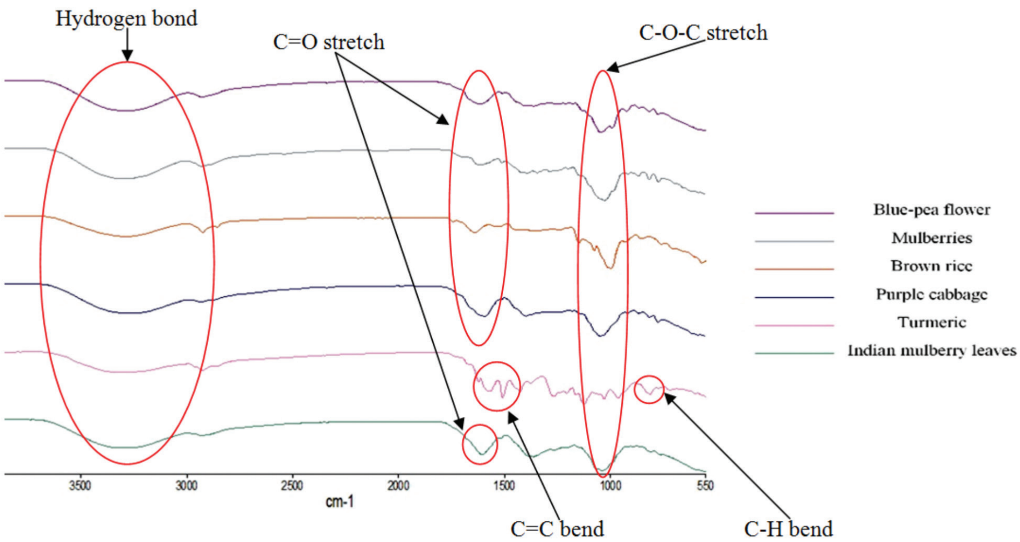

FIGURE 6. FTIR result for natural dyes

TABLE 3. Efficiency for all samples based on immersion time

Type of natural dyes	Immersion time, t (h)	Efficiency, η (%)
Blue-pea flower (Anthocyanin)	24	0.00073
	72	0.04400
	120	0.12300
Mulberries (Anthocyanin)	24	0.00377
	72	0.00570
	120	0.03200
Brown rice (Anthocyanin)	24	0.00066
	72	0.00484
	120	0.02040
Purple cabbage (Anthocyanin)	24	0.00023
	72	0.01950
	120	0.02000
Turmeric (Carotenoids)	24	0.00073
	72	0.03260
	120	0.03380
Mulberry leave (Chlorophyll)	24	/
	72	0.00019
	120	0.00200

The efficiency of turmeric dye was lower than blue-pea flower dye due to carotenoids molecule structure. Carotenoids were not being used as sensitizers for the DSCs for two reasons. First, most carotenoid species do not have effective functional group to bond with -OH of TiO_2. Secondly, strong steric hindrance of long chain alkane of carotenoid species prevents the dye molecules from arraying on TiO_2 film efficiently. Indian mulberry leaves contained chlorophyll molecular structure to produce the lowest efficiency as compared to other natural dye. The reason for such low efficiency was that the interaction between chlorophyll and TiO_2 was low. The structure of the pigment also affects the performance. For example, if the structure has longer R group, this result in the steric hindrance for the pigment to form bond with the oxide surface of the TiO_2 and hence prevent the molecule from arraying on TiO_2 film effectively. R group refers to a side chain. Hence, there was a lack of electron transfer from the dye molecule to the conduction band of TiO_2.

The highest efficiency of solar cell which was 120 h immersed in blue-pea solution was taken to be test with other electrolyte. The commercialized dye, N719 also has been tested with different type of electrolyte so that, comparison could be made. Gel electrolyte and solid electrolyte were used in order to see the best performance between three types of electrolyte. In general, Tables 4 and 5 show the efficiency of blue-pea flower sample and N719 sample.

Based on Tables 4 and 5, the result clearly showed that gel electrolyte has the highest efficiency for both samples. 0.203% of efficiency of gel electrolyte for Blue-pea flower dye and 0.024% of efficiency for N719 dye can be seen in Figure 7. Liquid electrolyte showed the second highest result of efficiency with 0.044% for Blue-pea flower and 0.017% for N719. For solid electrolyte, the efficiency obtained for Blue-pea flower and N719 were 0.005 and 0.003%, respectively. Although DSSCs based on liquid electrolytes have already achieved high conversion efficiencies, they also caused some substantial problems to put DSSCs into practical uses. For example, their high volatilities, solvent losses occur during long-term operations, resulting in decreases in DSSCs performances. The liquid electrolyte in cell presents several problems such as solvent evaporation and penetration of air and water caused by difficulty in long-term sealing especially in temperature cyclic tests (Kubo et al. 2001).

The gel polymer electrolyte has lots of advantages compared to other kind of charge transport material. Gel electrolyte contain high ioinic conductivity which were achieved by trapping a liquid electrolyte in polymer cages formed in a host matrix, good contacting and filling properties of the nanostructured electrode and counter electrode. Therefore, the gel polymer electrolytes have been attracting intensive attention. Up to the present, several types of gel electrolytes based on different kind of polymers have already been used in quasi-solid state dye-sensitized solar cells. Gel electrolyte exhibits the dependence of the ambient conductivity on EC (ethylene carbonate) content in the binary organic solvents mixture containing EC and PC (propylene carbonate). The amount of EC content has great influence on the ambient ionic conductivity of gel polymer electrolyte (Wu et al. 2006). Overall, the efficient transport of iodine and tri-iodine ions in the electrolyte is an essential requirement to guarantee the high performance of DSSC devices because the oxidized state of the dye must be regenerated efficiently by I^- ions after electrons from the excited state of the dye were injected into the conduction band of TiO_2. If the transport of iodine and tri-iodine ions is not efficient enough, the electrons in the conduction band of TiO_2 will recombine with the tri-iodine ions in the electrolyte, leading to a reduction in the electron current produced in the DSSC system.

CONCLUSION

In conclusion, blue-pea flower, turmeric, mulberries, brown rice, purple cabbage and Indian mulberry leave were successfully form on TiO_2mesoporous film using immersion method to produce TiO_2mesoporous

TABLE 4. The efficiency of natural dye sensitized solar cell from blue-pea flower

Electrolyte	J_{sc} (mA/cm^2)	V_{oc} (mV)	FF	η (%)
Liquid	0.390	0.205	0.545	0.044
Gel	0.680	0.480	0.622	0.203
Solid	0.024	0.450	0.481	0.005

TABLE 5. The efficiency of dye sensitized solar cell from N719 dye

Electrolyte	J_{sc} (mA/cm^2)	V_{oc} (mV)	FF	η (%)
Liquid	0.260	0.155	0.410	0.017
Gel	0.240	0.360	0.282	0.024
Solid	0.080	0.125	0.279	0.003

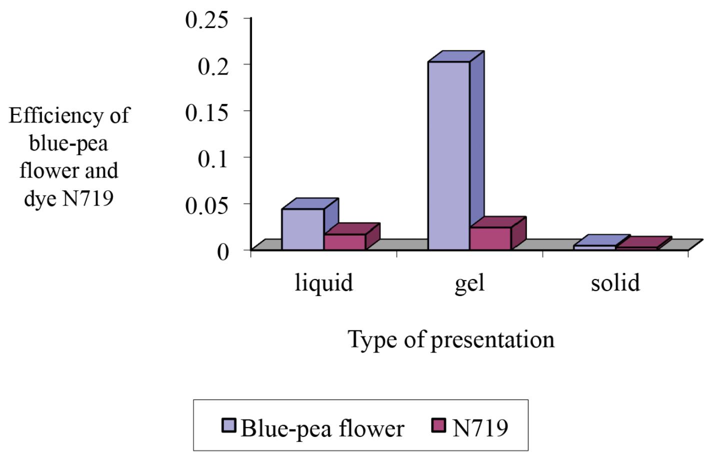

FIGURE 7. Efficiency of Blue-pea flower dye and N719 dye

photoanode for natural dye sensitized solar cells (DSSCs) assembly. Tests have been conducted in order to confirm the conductivity for all three types of electrolyte. Gel electrolyte showed high performance with 26.1 mS/cm of conductivity. However, liquid electrolyte and solid electrolyte obtained 17.34 and 0.45 mS/cm, respectively. Apart from physical characterization, the samples were subjected to electrical testing via current density-voltage (J-V) measurement. Blue-pea flower samples provided the highest efficiency. Due to carbonyl and hydroxyl groups presented on anthocyanin molecule, it can be bound with the surface of TiO_2 porous film, which was in favour of photoelectric conversion effect. Followed by turmeric, mulberries, brown rice, purple cabbage and lastly, Indian mulberry leaves. The differences in performances might be because of the different type of molecular structure of each natural dye. Another electrical testing has been done towards the sample of Blue-pea flower to identify the better performance of efficiency for liquid electrolyte, gel electrolyte and solid electrolyte. It was found that, gel electrolyte provided the highest efficiency compared to liquid electrolyte and solid electrolyte. Overall, the efficiency of each sample was obtained, but the efficiency obtained was quite low.

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