

Natural Dyes Extracted from Bioactive Components of Fruit Waste for Dye-Sensitized Solar Cell

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Abstract

DSSC using natural dyes extracted from bioactive components of red dragon fruit peel wastes were studied to determine the power conversion efficiency. Natural dyes were extracted by maceration method. Maceration is the soaking of fine powder from red dragon fruit peel for 24 hours in distilled water mixed with 10% citric acid with three variations of the material's weight ratio to the solvent, namely 1:4, 1:6 and 1:8. Materials were characterized by several tools such as SEM equipped with EDS, UV-Vis Spectrophotometry, and FTIR. The SEM analysis results with EDS showed that the TiO₂ thin layer had mesoporous morphology and showed Ti and O without other impurities with a composition of 73.79% oxygen (O) and 26.21% titanium (Ti). The results of UV-Vis characterization of the absorbance spectrum showed that there was absorption in the visible area, and the three samples of the dye solution had different absorbance power, photon energy, and absorption coefficient (α). FTIR analysis confirms that the sample shows a hydroxyl group, the building block for the anthocyanin structure. DSSC parameters were characterized using a solar simulator with 500 W/m² lighting. The best performance is obtained at a ratio of 1:8 with Voc = 0.47 V, Jsc = 23.46 μ Ac m⁻², Fill Factor = 0.480 and efficiency 0.029%

Keywords: Dye-sensitized solar cell, natural dye, TiO₂, FTO

I. INTRODUCTION

Renewable energy becomes a crucial thing in preserving nature and the environment quality [1]. Besides, it has also been proven to improve economic growth because of its lower cost than non-renewable energy [2]. Sunlight is one of the renewable energy that doesn't cause environmental pollution and noise [3]. Among all renewable technologies, solar cell installation, or commonly known as photovoltaic (PV), has dominated the renewable energy industry. Solar cell technology continues to develop because this technology is considered an environmentally friendly alternative energy source expected to replace fossil energy sources. The advancement of solar cell technology aims to produce cheap and high-efficiency solar cells [4].

The first generation of solar cells used crystalline silicon material. This technology can produce high-efficiency solar cells, but the production costs are quite expensive. This wafer-

based silicon solar cell panel still holds 90% of the worldwide PV production market [5].

Thin film technology is known as second-generation solar cells. Semiconductor materials are capable of producing thin-film cells, only a few micrometers thick [6]. The foremost materials in the manufacture of thin films are amorphous silicon (A-Si), cadmium telluride (CdTe), and copper indium gallium selenite (CIGS). This technology can be cheaper to produce but has a lower efficiency level than the first generation. In fact, this technology only contributes to 5% of the global PV market [7].

The third generation, namely Dye-Sensitized Solar Cell (DSSC), invented by O'Regan and Gratzel in 1991, has become a significant concern on solar energy because of its simple fabrication, good efficiency and low production costs. DSSC is different from silicon solar cells. In the use of DSSC technology, light absorption and cargo carrier transport is separated. The dye molecule is responsible for absorbing light while the charge separation is carried out by nanocrystalline inorganic semiconductors, which have a wide band gap such as TiO₂. In general, the DSSC has three main components. The three components are the working electrode, the counting electrode, and the electrolyte solution. The working electrode consists of transparent conductive glass such as Indium Tin Oxide (ITO) or Fluorine Tin Oxide (FTO), nanocrystalline TiO₂ semiconductor coating, and dye Counter electrode consists of transparent conductive glass that has been coated with carbon [8] or platinum [9]. The electrolyte used is an iodine trioxides electrolyte with a pair redox (I^- / I_3^-).

Sensitizers have a significant role in absorbing photons that come from sunlight and convert them into electrical energy. In the DSSC studies that have been carried out, synthetic dyes can achieve a conversion efficiency of 11-12% [10]. Sensitizers from synthetic dyes have been shown to provide better results in increasing the efficiency and durability of DSSC. However, it does show some drawbacks: more expensive and potentially lethal materials [11]. Some researchers are trying to use natural dyes instead of synthetic dyes to produce a less expensive and environmentally friendly sensitizer. The research that has been done uses leaves, fruits, roots, and mushrooms as the basic ingredients for natural dyes [12] [13] [14] [15] [16]. In this research, a DSSC study was conducted using bioactive components of red dragon fruit peel waste. It has the potential to be a natural dye because it produces a red color produced by pigments called anthocyanins such as cyanidin-3-sophoroside

and cyanidin-3-glucoside [17]. About 30-35% of dragon fruit is the peel, usually disposed of as waste that can pollute the environment. Thus, the study's objective is to identify the possibility of bioactive components of red dragon fruit peel waste for DSSC and determine the effect of variations in the material's weight ratio with the solvent. Dye solvents use distilled water because they are non-toxic, easy to obtain, and cheap. The use of these natural dyes can reduce the costs and increase added value compared to using pulp. Besides, fruit peel utilization can reduce the production of more and more waste due to increased red dragon fruit consumption for health.

II. MATERIALS AND METHODS

II.I Materials

The materials used in this study were FTO TCO22-7 glass with a material resistance of 7 ohm / sq and TiO₂ purchased from Solaronix SA. The TiO₂ thin layer measuring 6 x 6 mm was made using the screen printing technique on FTO glass measuring 20 x 20 mm and 2 mm thick. Red dragon fruit peel waste is used as a natural coloring agent.

II.II Extraction of natural dyes

The red dragon fruit peel is separated from the flesh and cut into small pieces. Then those samples were dried at room temperature in the laboratory. The dried sample was crushed in a blender to produce a fine powder. The dye solution was made by the maceration method. Maceration is soaking the fine powder of red dragon fruit peel for 24 hours in distilled water mixed with 10% citric acid, with three variations of the material's weight ratio to the solvent, namely 1:4 1:6 and 1: 8. The extract is filtered with filter paper and accommodated in bottles.

II.III DSSC assembly

The making of working electrodes begins by immersing the conductive glass coated with TiO₂ for 24 hours in the three variations of the solvent ratio. The Counter Electrode is made of platinum-coated FTO conductive glass. Making DSSC with a sandwich system, where the working electrodes are stacked with a counter electrode and electrolyte solution is dropped between the two electrodes (Iodolite HI-30). The two sides are clamped with a binder clip.

II.IV Characterization

The TiO₂ thin layer on FTO glass was characterized by SEM equipped with EDS (JEOL-JSM-6510LA). The dye solution's characterization was carried out using Fourier Transform Infrared Spectrophotometer IRPrestige-21 and UV-Vis Spectrophotometry (UV-Vis-1800 Shimadzu). The dye band gap's determination absorbed by the TiO₂ surface is calculated using the formula (1). Where h is Planck's constant, ν is the frequency, λ is the wavelength, and c is the light speed.

$$\text{Photon Energy } (E) = h \cdot \nu = \frac{h \cdot c}{\lambda} \quad (1)$$

The absorption coefficient determines the extent to which a material can penetrate a particular wavelength of light before being absorbed. The absorbance coefficient for each wavelength is obtained by dividing the absorbance by the wavelength shown in the formula (2) using Boltzmann's K constant [18].

$$\text{absorption coefficient } (\alpha) = \frac{4\pi k}{\lambda} \quad (2)$$

The three DSSC samples that have been made were characterized by the Oriel Solar Simulator Model 81193 and the I-V meter Keithley 2400 Source Meter (SMU) instrument at the Electronics and Telecommunications Research Center, Indonesian Institute of Sciences (P2ET-LIPI) Bandung. The short circuit current I_{sc} or J_{sc} and the open-circuit voltage V_{oc} are determined from the I-V curve. J_{sc} is the current density, i.e., I_{sc} divided by the area of the active cell. The maximum output power (P_{max}) is determined when the product and voltage are maximum. P_{max} can be calculated by equation (3).

$$P_{max} = V_{mp} \cdot I_{mp} = V_{mp} \cdot J_{mp} \quad (3)$$

I_{mp} and V_{mp} are the values of current and voltage at maximum power, respectively. The fill factor (FF) measures the device's ideality and is defined as the maximum power output ratio per unit area to the V_{oc} and I_{sc} products. The fill factor (FF) is calculated by equation (4).

$$FF = \frac{V_{mp} \cdot I_{mp}}{V_{oc} \cdot I_{sc}} = \frac{V_{mp} \cdot J_{mp}}{V_{oc} \cdot J_{sc}} \quad (4)$$

The overall efficiency of converting solar energy to solar cell electricity is defined as the maximum cell output ratio divided by incident light power. This can be determined by the measured photo current density at short circuit (I_{sc}), open-circuit voltage (V_{oc}), Fill Factor (FF), and incident light intensity (P_{in}), incident irradiation power (E_{in}), active cell surface area. (A) [19], as shown in equation (5) below.

$$\eta = \frac{P_{max}}{P_{in}} = \frac{V_{mp} \cdot I_{mp}}{E_{in} \cdot A} = \frac{V_{oc} \cdot I_{sc} \cdot FF}{E_{in} \cdot A} = \frac{V_{oc} \cdot J_{sc} \cdot FF}{E_{in} \cdot A} \quad (5)$$

III. RESULTS AND DISCUSSION

Scanning Electron Microscope (SEM) is an electron microscope used to directly investigate solid objects' surface. The characterization of TiO₂ thin films to see the morphology was carried out by SEM JEOL JSM-6510 LA at an acceleration voltage of 15 kV and magnification of 25,000 times, and the results are as shown in **Fig. 1**.

In general, the particles produced are mesoporous and are on the micrometer size scale. In addition to the TiO₂ thin film's morphology, it was also characterized by Energy Dispersive Spectroscopy (EDS). EDS is an analytical technique for analyzing the elements or characteristics of a chemical object. EDS is used to verify titanium and oxygen content and the absence of other contaminants to maintain sample quality. The TiO₂ thin layer's analysis results on FTO TCO22-7 glass with

EDS (JEOL-JSM-6510LA) at an acceleration voltage of 20 kV, a probe current of 7.47500 nA and a magnification of 3,000 times is shown in **Fig. 2**.

The investigation results show the presence of Ti and O without other impurities with a composition of 73.79% oxygen (O) and 26.21% titanium (Ti).

Optical characterization of the three red dragon fruit peel dye solution samples using a UV-Vis (UV-1800 Shimadzu) spectrophotometer in the wavelength range of 400-800 nm, visible light spectrum. Within this wavelength range, organic dyes are naturally effective in absorbing light in visible light during the photosynthesis process. The results of the absorbance spectrum characterization are shown in **Fig.3**.

The three sample solutions have different absorption peaks. The absorption peak at this wavelength indicates the presence of anthocyanin content in the dye. The highest absorption peak was obtained at a ratio of 1: 4, namely 0.587, at a wavelength of 533.50 nm. All the pigments can absorb photons of light and generate excited electrons. The process allows a TiO₂ conduction band with acceptable performance to increase the DSSC conversion efficiency [20]. Dyes containing anthocyanin pigments have sufficient hydroxyl groups to attach TiO₂ firmly and can inject electrons into the TiO₂ conduction band when excited by visible light [21]. Low efficiency is the major problem in DSSC. The cause is the weak bond between the dye molecule and TiO₂, whose electrons can be carried from the excited state molecule in the TiO₂ film [22]. So that the use of dyes from dragon fruit peel can increase the interface between the sensitizers and the molecules of TiO₂, which are interconnected to improve the efficiency of DSSC conversion. The energy discrepancy between the conduction band and the valence band is known as the band gap energy, which in DSSC performance is related to the absorbed solar energy. The results of the three samples' characterization of the absorbance spectrum have differences in photon energy and absorption coefficient (α), as in **Table 1**.

The dye solution was characterized by Fourier Transform Infrared (FTIR) Spectroscopy Prestige 21 produced by Shimadzu Japan from a wavenumber 400-4000 cm⁻¹, and the results are as shown in **Fig. 4**.

The absorption peak in the infrared spectrum indicates that the absorption is related to the functional groups contained in the dye. The OH functional groups are identified in wave numbers 3651.25 cm⁻¹, up to 2872.01 cm⁻¹, as shown in **Table 2** [23].

The O-H functional group is a hydroxyl group that makes up the anthocyanin structure. The hydroxyl group functions as an adhesive for the pigment compound and TiO₂ layer on the FTO. Red dragon fruit peel dye molecules containing hydroxyl and carbonyl groups prove the interaction between TiO₂ and dye as functional groups that can interact on the TiO₂ surface [24], which will contribute to the efficiency of photovoltaic conversion [25].

In this research, red dragon fruit peel waste was successfully used to make natural dyes in the DSSC prototype. **Table 3** summarizes the short-circuit current, open-circuit voltage, fill factor, and efficiency extracted from the I-V curve.

The voltage and current output results obtained using the Solar Simulator AM 1.5 light source and equipped with a pyranometer sensor with an Intensity of 500 W/m² show that the resulting voltage is appropriate and constant. Still, the resulting current is not ideal [26]. The voltage on the DSSC-based solar cell is generated from the distinction of the conduction energy level of the TiO₂ semiconductor electrode with the electrochemical potential of the resulting current redox electrolyte pair concerning the number of photons involved in the conversion process. Overall, the flow obtained is still small. The small output current generated is due to the very large resistance of the TiO₂ semiconductor electrode layer and electrolyte due to the fast drying electrolyte. The main factor affecting resistance is electrolytes [27]. With this very large resistance value, the dye's injected electrons experience a very large resistance in the TiO₂ layer. It makes the number of electrons flowing into the circuit the outermost becomes small.

Another cause can be caused by the dye's inadequate function in forming and injecting electrons into the TiO₂ electrode layer. DSSC efficiency is influenced by three parameters, namely, I_{sc} (short circuit current), V_{oc} (open circuit voltage), and FF (Fill Factor). I_{sc} is a short circuit that occurs when the voltage can equal zero. This current is equal to the number of photons converted into electron-hole pairs. The more electrons are excited, the greater the resulting efficiency. V_{oc} is the voltage value when the current is zero because all the excitons are combined so that no current flows in the DSSC. FF is the ratio of maximum power (P_{max}) to short circuit current (I_{sc}) and open-circuit voltage (V_{oc}). Several factors can influence FF. One of them is high internal resistance that causes low fill factors and decreased efficiency. The increase in electron mobility affects the increase in the charging factor. Increasing electron mobility increases current. The best performance is obtained at a ratio of 1:8 with V_{oc} = 0.47 V, J_{sc} = 23.46 μAcm^{-2} , Fill Factor = 0.480 and efficiency 0.029%. These results prove that natural dyes made from the use of red dragon fruit peel waste can be used as a sensitizer. However, further research to improve the DSSC efficiency through optimization of current and voltage is necessary.

IV. CONCLUSION

An investigation was carried out to determine DSSC power conversion efficiency using natural dyes extracted from bioactive components of red dragon fruit peel waste. SEM analysis results equipped with EDS showed that the TiO₂ thin layer had mesoporous morphology and Ti and O's presence without other impurities. The composition of 73.79% oxygen (O) and 26.21% titanium (Ti). The results of UV-Vis characterization of the absorbance spectrum showed that there was absorption in the visible area, and the three samples of the dye solution had different absorbance power, photon energy, and absorption coefficient (α). FTIR analysis confirms that the sample shows a hydroxyl group, the building block for the anthocyanin structure. This hydroxyl group is suitable as an adhesive between pigment compounds and a thin layer of TiO₂ on FTO. The created DSSC parameters are determined under 500 W/m² lighting. It is known that the voltage generated is good, but the current obtained is still small. The small output

current generated is due to the very large resistance of the TiO₂ semiconductor electrode layer and electrolyte due to the fast drying electrolyte. The best performance is obtained at a ratio of 1:8, with an efficiency of 0.029%. Although the efficiencies

obtained are still low, the results are encouraging and could lead to additional research-oriented towards finding new, naturally sensitive substances.

Table 1. Photon energy and absorption coefficient (α) of natural dye from red dragon fruit peel waste

Ratio	Wavelength (nm)	Peak Absorbance	Photon Energy (eV)	Absorption coefficient (α) (km ⁻¹)
1:4	533.50	0,587	2.33	2.029
1:6	541.00	0,313	2.30	2.001
1:8	535.50	0,233	2.32	2.021

Table 2. FTIR spectrum peaks in red dragon fruit peel dye solution

Ratio 1:4		Ratio 1:6		Ratio 1:8	
Peak (cm ⁻¹)	Intensity (%)	Peak (cm ⁻¹)	Intensity (%)	Peak (cm ⁻¹)	Intensity (%)
416.620	50.176	449.410	56.717	455.200	62.581
526.570	54.208	528.500	57.846	522.710	61.953
858.320	22.443	891.110	21.874	860.250	23.695
1130.290	30.608	1132.210	30.135	1128.360	31.930
1394.530	32.021	1394.530	31.447	1249.870	30.723
1508.330	31.759	1412.190	31.107	1394.530	32.976
1695.430	27.056	1695.430	26.212	1514.120	32.520
2376.300	35.052	1774.510	26.523	1697.360	27.223
2872.010	47.418	2378.230	34.465	2382.090	35.673
2997.380	47.358	2872.010	48.032	2875.860	48.574
3278.990	56.402	3261.630	57.654	2989.660	48.940
3651.250	17.737	3651.250	16.127	3622.320	17.409

Table 3. DSSC performance is based on natural dyes extracted from red dragon fruit peel waste with various solvent ratios

Ratio	V _{oc} (V)	J _{sc} (μAcm ⁻²)	V _{mp} (V)	J _{mp} (μAcm ⁻²)	P _{in} (Wcm ⁻²)	P _{max} (μWcm ⁻²)	Fill Factor	Efficiency (%)
1:4	0.26	19.74	0.15	10.23	0.018	1.534	0.299	0.009
1:6	0.40	23.60	0.28	14.96	0.018	4.189	0.444	0.023
1:8	0.47	23.46	0.33	16.05	0.018	5.295	0.480	0.029

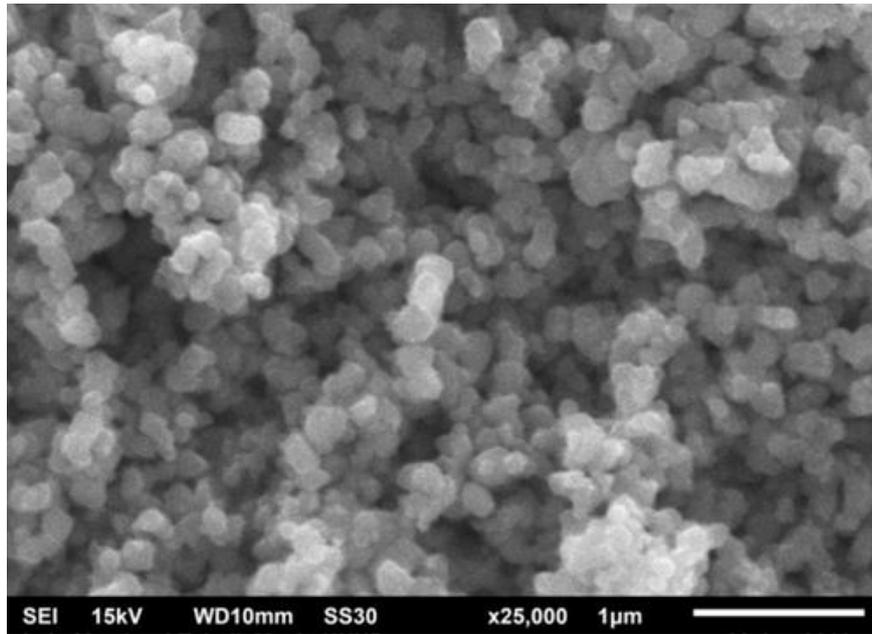
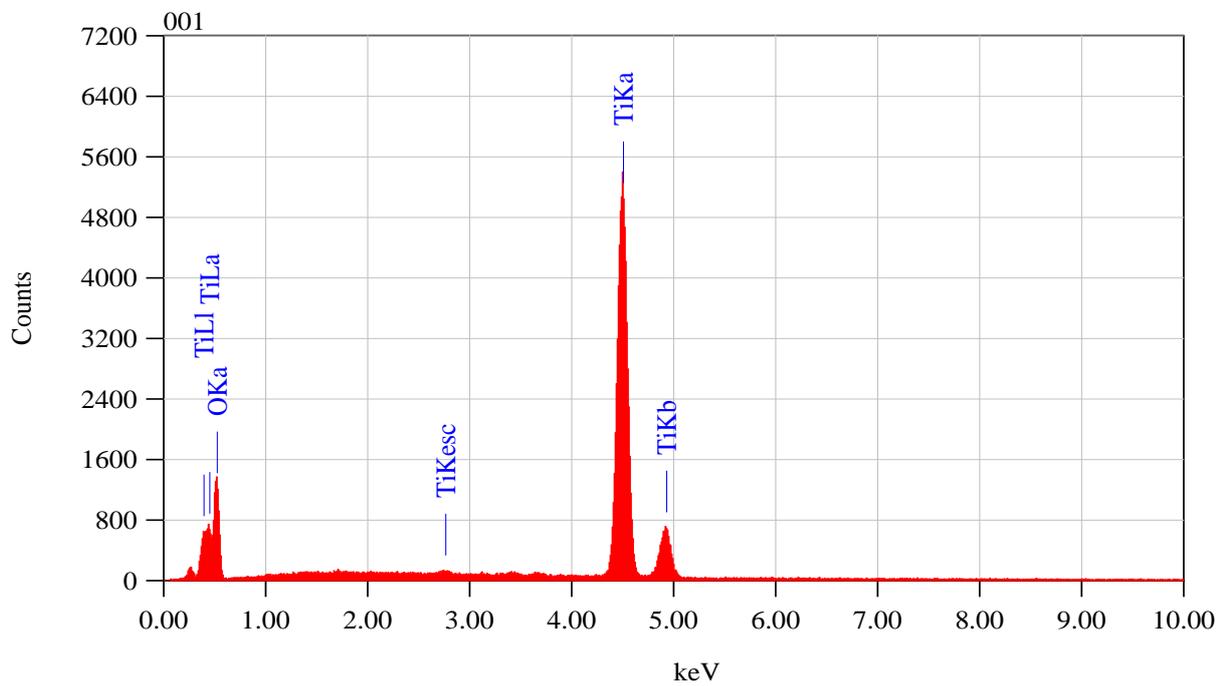


Fig. 1. Characterization of the TiO₂ thin film morphology by SEM



ZAF Method Standard less Quantitative Analysis

Fitting Coefficient : 0.2158

Element	(keV)	Mass%	Sigma	Atom%	Compound	Mass%	Cation	K
O K	0.525	48.46	0.58	73.79				24.9038
Ti K	4.508	51.54	0.27	26.21				75.0963
Total		100.00		100.00				

Fig. 2. Graph of TiO₂ thin film element analysis

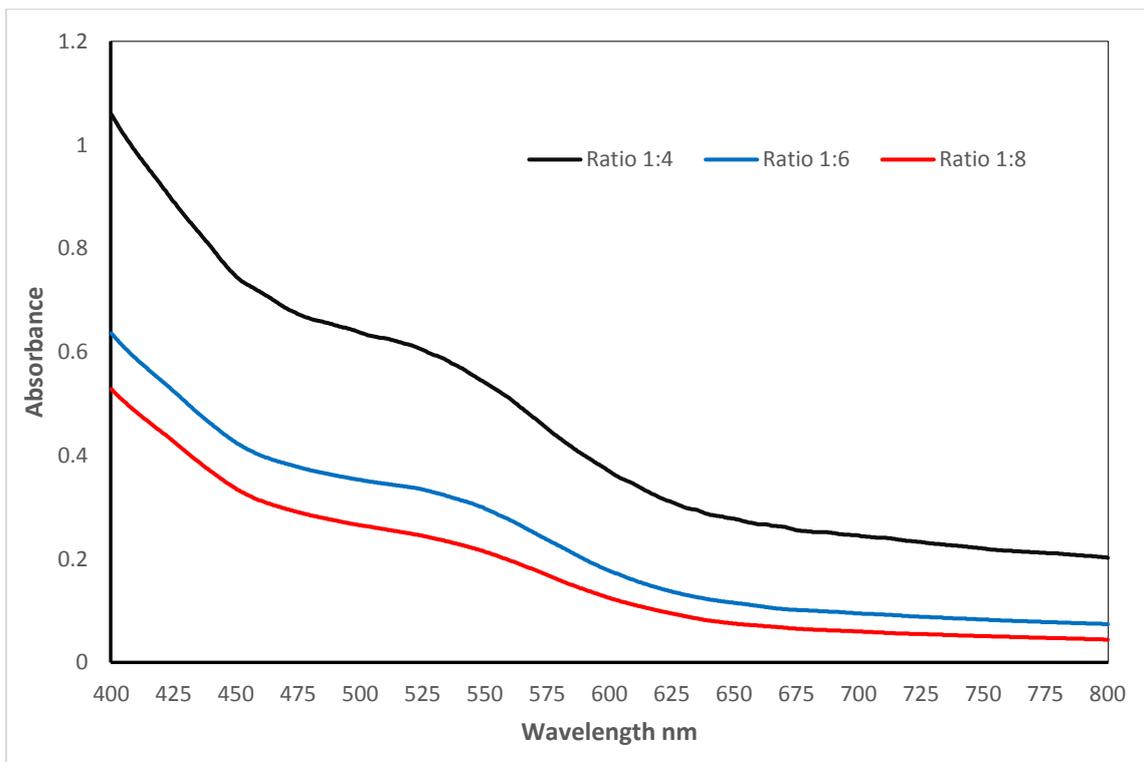


Fig. 3. UV-Vis spectrum of red dragon fruit peel dye

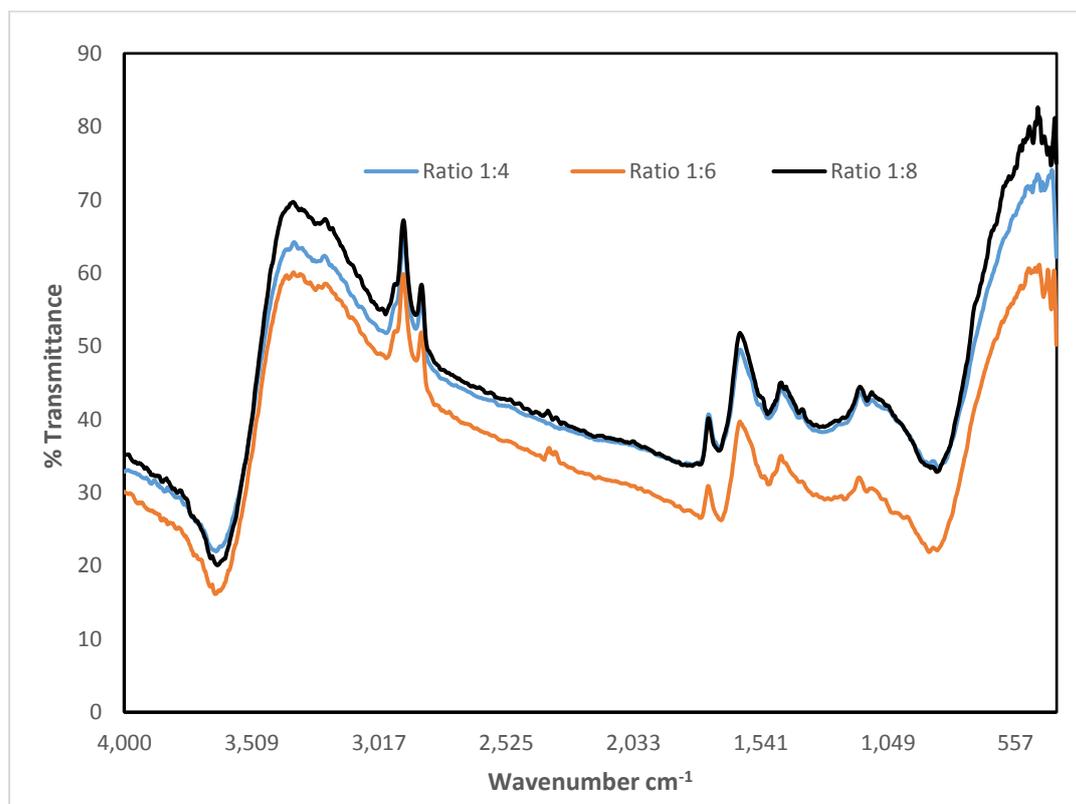


Fig. 4. The results of FTIR analysis of red dragon fruit peel with various solvent ratios

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