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Effect of Natural Dye Extracting Solvents on the Performance of Dye-Sensitised Solar Cells

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Abstract. Dye-sensitised solar cells (DSSCs) have attracted immense attention in photovoltaics research and development due to the low cost of fabrication, sustainability and use of abundant natural resources. Natural dyes were adapted to DSSCs as a sensitiser but unfortunately proved to have poor efficiency. Herein, we manufactured two organic dyes from yellow and red gambier fruits and treated both dyes with methanol and isopropyl alcohol. The photoanode and photocathode were prepared from TiO_2, sensitiser and indium thin dioxide (ITO); and carbon and indium thin dioxide, respectively. An electrolyte was made by mixing potassium iodide and iodine. The doctor blade method was used to add the semiconductor to the surface of the conducting ITO. UV–Vis and FTIR spectroscopic measurements have been performed to determine the absorbance and functional groups in the gambier extracts. From the analysis of UV–Vis and FTIR spectra, and photovoltaic measurements made under solar irradiation, we found that the gambier extracts are dominated by tannin and flavonoids and that dyes using isopropyl alcohol as the solvent have higher efficiency than using methanol, respectively. This indicates that the gambier extracts and solvents might influence the performance of DSSCs.

1. Introduction

Photovoltaics, or solar cells, is a technology which converts solar energy into electricity, which is renewable and sustainable. At present, there are three generations of solar cells: the first and second are single-multi-silicon and thin-film solar cells, but unfortunately, these are expensive to fabricate [1]. Dye-sensitiser solar cells (DSSCs) are the next generation and have attracted tremendous attention because of their low cost and eco-friendly production, use of readily available raw materials, and ease of fabrication [2]. DSSCs consist of three crucial components: the photoanode, the photocathode and the electrolyte. Simply, a glass conductor is coated with a thin film of semiconductor material and immersed in a sensitiser dye, and the DSSC is completed with a platinum counter electrode and I^-/I_3^- as the electrolyte. Then, the solar energy is converted to electricity by the absorption of photons by the dye molecule, supplying energy to electrons injected into the conduction band of the semiconductor materials [4].

Generally, the dye as a sensitiser in solar cells plays the important role of absorbing photons, and the best dye molecules can capture many more photons. Additionally, there are two kinds of dyes which are frequently applied in DSCCs, called synthetic and organic dyes. The synthetic dyes such as ruthenium [5][6] and N719 [7], have higher efficiency than organic dyes, but, unfortunately, they are expensive and require special treatment. Recently, numerous researchers have developed sensitisers from the flowers, woods, fruits and leaves of organic dyes which are abundant and inexpensive resources and

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consist of pigments such as flavonoids, anthocyanins, carotenoids, and tannins [3][8]. According to Nasyori et al. they applied anthocyanin from *sappan linn* wood which gained efficiency (0.196 %) [9], mango (0.43 %) [10], *rosella* (0.37 %) [11], pawpaw leaf (0.2 %) [12], indigo core [13], and *pterocarpus indicus willd* leaf (0.03 %)[14]. Nevertheless, the low efficiency is still a challenge; numerous improvements to them have been tried, such as adding other materials in the dye, varying the concentration, and changing the macerating technique.

With regard to this challenge, we developed a new dye from gambier extracts and modified the maceration process by using the solvents isopropyl alcohol and methanol on each gambier extract. We discovered that the gambier extracts could be employed successfully as a sensitiser and investigated the use of both solvents to improve the performance of the DSSCs.

2. Materials and methods

2.1. Preparation of DSSCs

The titanium dioxide (TiO_2) and indium tin oxide (conducting ITO) were purchased from Titanos (Shanghai) and Latech Scientific (Singapore) with the specifications of TiO_2 (99%, 5–10 nm) and 7–10 Ω/sq and 20 × 20 × 7 mm, respectively. After that, the basic ingredients were obtained locally from Indonesia such as yellow and red gambier fruits (West Sumatra); methanol and isopropyl alcohol (99%); KI and I_2 (99%, solid, ROFA), and a candle.

2.2. Preparation and extraction of natural dyes

Firstly, the sensitiser was prepared from the natural dyes of the yellow and red gambier fruits. Then, the samples were rinsed for 5 minutes in aquabidest distilled water and dried at room temperature for 2 h. Secondly, the yellow and red gambier fruits were divided into 200 g aliquots and macerated in 300 ml each of methanol and isopropyl alcohol for 24 h. Then the samples were evaporated for 3 h and kept at room temperature for 2 h. Finally, the resulting samples were divided and measured using an Agilent 8453 UV–Visible spectrophotometer and a Shimadzu IR prestige-21 FTIR spectrometer.

2.3. Fabrication of DSSCs

To make the photoanode, first, 2 g of the pure TiO_2 was weighed and separately mixed with 3.2 ml each of methanol and isopropyl alcohol, for all samples. Meanwhile, the conducting ITO was prepared, its resistivity was measured, and it was washed in methanol and isopropyl alcohol in an ultrasonic cleaner for 8 minutes. Then it was placed on tissue at room temperature for 20 minutes. The mixtures of TiO_2 and methanol and isopropyl alcohol were doped on the ITO-positive surface using the doctor blade method in a 1 cm^2 area. Having doped the conducting glass with TiO_2, it was inserted into the oven at 200 °C for 90 minutes and then kept at room temperature for 90 minutes. Finally, the TiO_2 film was immersed in the previously-prepared dye concentrations for 24 h. To create the photocathode, conducting ITO was prepared, its resistivity was measured, and it was washed with methanol and isopropyl alcohol in an ultrasonic cleaner for 20 minutes, then it was placed on the tissue at room temperature for 30 minutes. Then, the positive area of the ITO was burned with the candle, and a 1 cm^2 area was made on the ITO glass. Finally, to make the electrolyte, 5 g of solid KI and 0.5 g of I_2 were prepared and mixed with aquabidest distilled water in the ultrasonic cleaner three times for 20 minutes each.

2.4. Photocurrent and photovoltage of DSSCs

After the three important components were created, the working electrode and counter electrode were assembled into a sandwich, and the doped electrolyte was placed in both electrodes. The measurements were carried out in full sunlight in West Sumatra from 10.30 – 12.00 (GMT+7) using a potentiometer (10k) and a light meter to measure input power. The setup of photocurrent and photovoltage measurements and the cell illustration are shown in Figure 1.

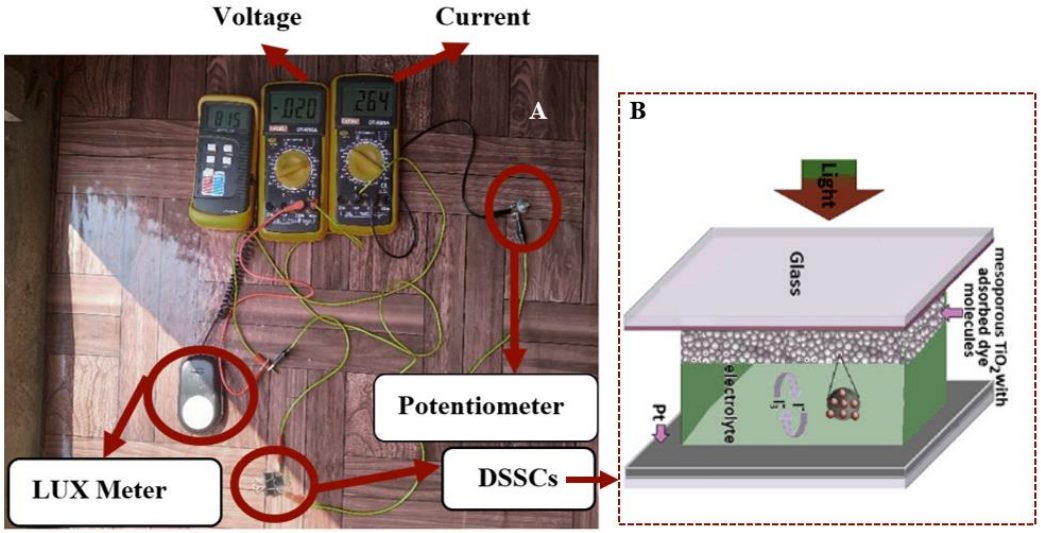


Figure 1. (A) Experimental setup for photocurrent and photovoltage measurements and (B) structure illustration of DSSCs.

3. Results and discussion

3.1. UV–Vis spectra

After the yellow and red gambier fruits were macerated and evaporated, the gambier extracts were immersed in two solvents (isopropyl alcohol and methanol) and characterised using UV–Vis spectroscopy in the range 300–800 nm to analyse the absorbance and wavelength. Figure 2 and 3 show the gained absorbance and calculated energy bandgap in each gambier extract, respectively.

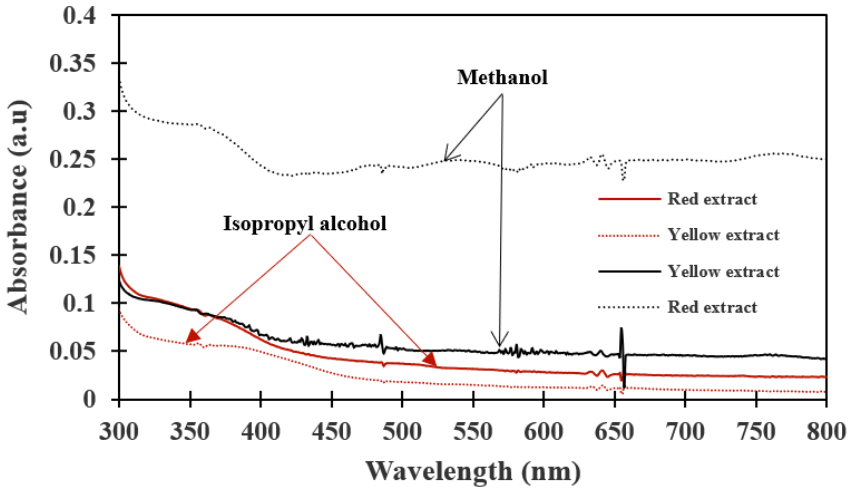


Figure 2. Characteristic of the gambier extract prepared in methanol (black solid and dashed lines) and that prepared in isopropyl alcohol (red solid and dashed lines) examined by UV–Vis spectrophotometer.

Generally, in the UV–Vis measurements in Figure 2, many peaks appeared in the ranges 638–655 nm and 486–657 nm in each gambier extract in isopropyl alcohol and methanol. It is apparent that gambier extract is dominated by tannin and flavonoid which is commonly found out in fruit and wood and possesses a characterization of yellow pigment. Figure 3 illustrates the bandgap energies which have been calculated. The yellow and red gambier dyes were found to have bandgap energies of 2.69 and 2.90 eV for isopropyl alcohol and 2.61 and 2.81 eV for methanol solvents, respectively. A low bandgap

in organic dye as a photosensitizer has a crucial rule to trigger electrons moving faster to the semiconductor. It is evident that a wide bandgap will retard electrons and decrease the performance of DSSCs.

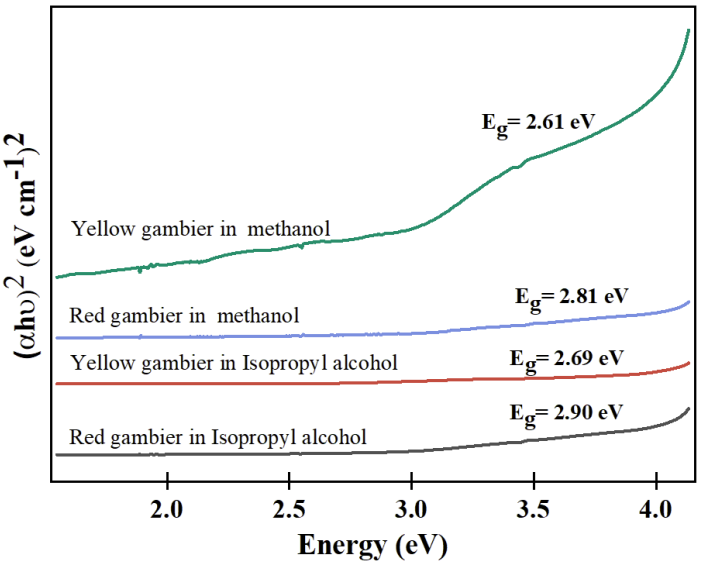


Figure 3. Bandgap energies of gambier extracts in methanol and isopropyl alcohol solvents

3.2. FTIR characteristics

The gambier extracts were characterised by FTIR in the range 1000–3000 cm^{-1} to identify functional groups and the contents of the gambier extracts. Figure 4 shows that many peaks appeared having different transmittances and at different wavenumbers in each gambier extract. Herein, the investigation of gambier extracts showed that the functional groups in the 1600–1400, 1300, and 1190–1000 cm^{-1} ranges are C–C, –CH_3, and C–O, respectively [15]. Additionally, the researchers [16][17], revealed that the compounds in the gambier are dominated by tannin and flavonoid, which have the potential to be the sensitiser in dye-sensitised solar cells [3].

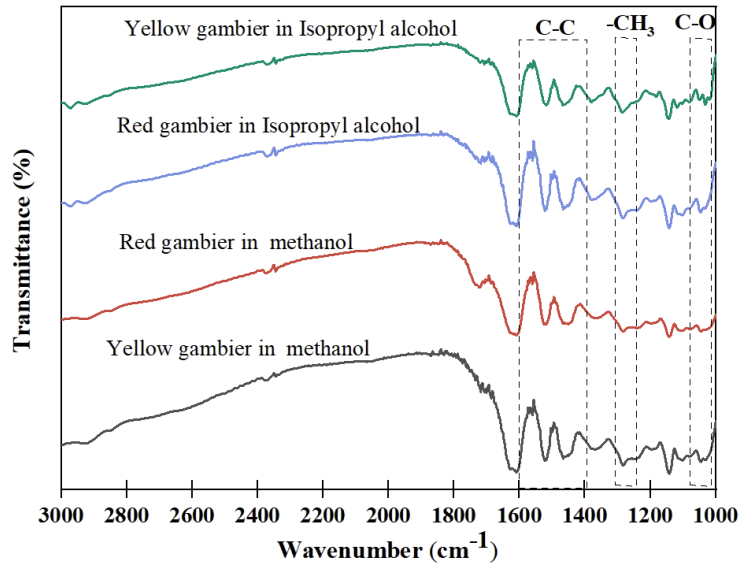


Figure 4. Characteristics of gambier extracts determined by FITR spectroscopy

3.3. *Photocurrent and photovoltage of DSSCs*

Characterisation of the photocurrent and photovoltage is a pathway to analysing the ability of DSSCs to convert solar energy to electricity. Previously, the photoanode was immersed in gambier extracts prepared with isopropyl alcohol and methanol for 24 h, and the photocathode was doped by the candle, they were assembled in a sandwich, and then the electrolyte was injected in both electrodes. Then, the measurement of the photocurrent and photovoltage was performed under outdoor solar irradiation, as shown in Figure 1. Here, a lux meter was used to detect the input photocurrent in the DSSCs. The photocurrent-photovoltage, fill factor and efficiency were calculated using Equations (1) and (2) below

$$FF = \frac{V_{max} J_{max}}{V_{oc} J_{sc}} \tag{1}$$

$$\eta = \frac{J_{sc} \times V_{oc} \times FF}{Pin} \times 100\% \tag{2}$$

Where FF is the fill factor, V_{oc} and V_{max} are the highest and maximum photo voltages (V), J_{sc} and J_{max} are highest and maximum photocurrent (mA/cm^2), P_{in} and P_{out} are input and output energy (mW/cm^2), and η is the efficiency (%).

Table 1. Photocurrent and photovoltage of DSSCs

Solvent	Sample	J_{max} mA/cm^2	V_{max} Volt	J_{sc} mA/cm^2	V_{oc} Volt	FF	P_{in}	%
Isopropyl alcohol	Yellow Gambier	0.643	0.300	1.181	0.568	0.288	10	2.0185
	Red Gambier	0.688	0.309	0.978	0.656	0.331	10	2.2245
Methanol	Yellow Gambier	0.699	0.254	0.702	0.469	0.539	10	1.8578
	Red Gambier	0.469	0.293	0.752	0.624	0.293	10	1.4379

Table 1 and Figure 5 show that the photocurrent and photovoltage have been recorded successfully. In Table 1, it is evident that the highest efficiency is 2.2% and the next highest is 2.0% in the yellow and red gambier extracts in isopropyl alcohol, respectively. The J_{max} and J_{sc}, V_{max} and V_{oc}, and FF are 0.688 and 1.181 mA/cm^2, 0.309 and 0.656 V, and 0.331 for the dyes prepared with isopropyl alcohol. The efficiency of DSSCs with yellow and red gambier extracts prepared with methanol are 1.8 and 1.4 %. The J_{max} and J_{sc}, V_{max} and V_{oc}, and FF were obtained as 0.469 and 0.702 mA/cm^2, 0.254 and 0.469 V, and 0.293. In this regard, the gambier extract prepared in isopropyl alcohol is more efficient and has better behaviour as DSSCs than that prepared in methanol.

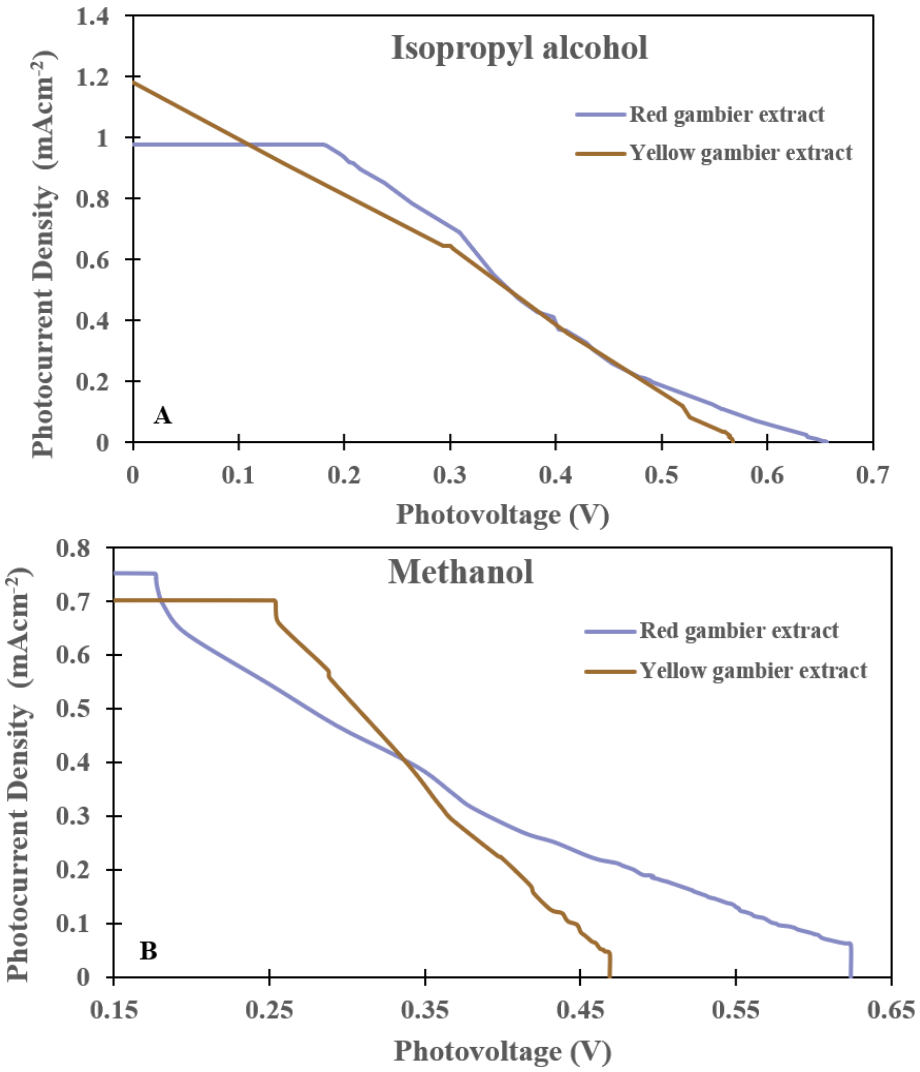


Figure 5. Photocurrent density vs. photovoltage curves of DSSCs prepared using gambier extracts (A) isopropyl alcohol (B) methanol

Figure 5 depicts the curves of photocurrent and photovoltage measured when using natural dyes from yellow and red gambier extracts prepared in isopropyl alcohol and methanol. As can be seen that the yellow gambier extract of isopropyl alcohol has attained the highest J_{sc} which is approximately 1.2 mA/cm^2. While the gambier extract immersed by methanol, merely shows from 0.7 to 0.75 mA/cm^2 of J_{sc}. This research indicates that the solvent used in the preparation of organic dye is crucial to improving the performance of dye-sensitised solar cells.

4. Conclusion

The natural dye has an important role as a sensitiser in dye-sensitised solar cells. Here, we utilised new natural dyes made from yellow and red gambier fruit extracts and prepared using two solvents, isopropyl alcohol and methanol. From the characterisation of the dyes by UV–Vis and FTIR spectroscopy, numerous peaks appeared in the range 486–657 nm in both extracts and solvents, and tannins and flavonoids dominate the compositions of the dyes. The measurement of the photocurrent and photovoltage revealed that organic dyes from gambier extracts have excellent ability as a sensitiser in DSSCs. Additionally, the highest efficiency (2.2%) was obtained by the red gambier in isopropyl alcohol, and the next highest efficiency is 2.0% obtained in yellow gambier extracts immersed in isopropyl alcohol.

Acknowledgement
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