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Analysis of Photodegradation and Reaction Kinetics of Textile Organic Dyes on *Caesalpinia Sappan, Lawsonia Inermis* and *Areca Catheu* using Spectroscopic Methods for Solar Cell Application

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ABSTRACT

Organic dyes are part of the textile industry that have a negative impact on the environment. Photodegradation using sunlight can reduce the environmental impact as it has the potential to convert complex organic dyes into simpler and less harmful compounds. Natural dye-based hybrid solar cells have the lowest cost among solar cell technologies, gives the lowest photochemical conversion efficiency. The problematic factor in Dye-Sensitised Solar Cells (DSSC) is the use of electrolyte type, either gel or solution, which lead to the leakage, dye instability, and evaporation. The aim was to analyse the photodegradation reaction kinetics of organic dyes in *Caessalpinia Sappan, Medina Lawsonia Inermis, Yemeni Lawsonia Inermis* and *Areca Catheu* using spectroscopic methods. This research focuses on the potential of natural dyes extracted from these sources as sensitizers in DSSC. The stability of natural dye was analysed using the photodegradation reaction kinetics of organic dyes before and after treatment using advanced spectroscopic methods. The extracted natural dyes were determined their optical absorption and functional group through Ultra Violet-Visible (UV-Vis) spectrophotometer, UV-Vis and Fourier Transform Infrared Spectroscopy (FTIR), respectively. The photo degradation proses of natural dyes were done under radiation of sunlight for four weeks with measured their photodegradation efficiency at every weeks. After plasma treatment using APPJ, a positive effect on the stability and colour of anthocyanins was observed. The results showed an increase in the percentage of photodegradation after APPJ treatment on *Medina Lawsonia Inermis* and *Yemeni Lawsonia Inermis*. While *Caessalpinia Sappan*, and *Areca Catheu* showed a slight change compared to before plasma treatment. The FTIR spectra of the natural dyes showed the presence of functionalised hydroxyl compounds and C-O groups. After plasma treatment, stronger functional group peaks were observed and new chemical bonds were formed.

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1. Introduction

The world is currently experiencing global warming, which results in abnormal weather conditions such as erratic rainfall, floods, heat waves, tropical cyclones, etc. Efforts are needed to save the earth by preserving natural resources and reducing CO_2 emissions to protect the environment. The technology to produce hydrogen ecologically and minimize CO_2 emissions is through Renewable Energy Sources (RES). Solar energy generation is a renewable energy source that is environmentally friendly and energy-efficient [1,2]. The electromagnetic wave spectrum demonstrates the wavelength and intensity of light, and dark colors absorb more solar heat than light colors [3].

Recently, it has been necessary to develop renewable energy that is low-cost, environmentally friendly, and has no negative effects on humans. The cost of hybrid solar cells based on natural dyes would be the lowest among all solar cell technology, however, the power-conversion efficiency is also the lowest. Therefore, it is urgent to increase the power conversion efficiency of hybrid solar cells based on natural dyes for future competition.

The disadvantages of Organic Photovoltaic (OPV) are low efficiency and short service life. A Dye-Sensitized Solar Cell (DSSC) and Perovskite Solar Cell (PSC) are the type of third-generation solar cells that uses photoelectrochemical principles [4,5]. This type of solar cell is believed to provide alternative energy concepts with more affordable production costs and simpler fabrication technology. DSSC is assembled using natural dyes from leaves with chlorophyll as a sensitizer [6-8]. The absorbance spectrum of anthocyanins is in the visible range (450-580 nm), and its pigments are red, blue, and purple in fruits, flowers, and leaves [9,10]. Photoanode such as titanium dioxide thickness or layer also could be an important parameter to enhance the performance of DSSCs [11,12]. Aid *et al.*, discussed various types of plant extract for this type of hybrid photovoltaic system [13].

Some countries impose strict environmental regulations in response to the hazardous waste generated by synthetics due to non-consumable dyes of varying severity. Hence, the interest in experimenting with natural dyes is increasing. Synthetic experimentation of natural dyes will result in dyes whose use is environmentally friendly, safer, non-toxic, and renewable [14].

One factor that is still a problem and weakness in DSSC is electrolyte types, both gels, and solutions, where because the form is generally in the form of a solution, that could cause a leakage. This is due to anthocyanin instability in the dye, evaporation, and the possibility of corrosion on the counter-electrode [15,16]. Thus, in this research, the Atmospheric Pressure Plasma Jet (APPJ) method will be used to treat the natural dye. Previous studies have found that plasma treatment with APPJ increases anthocyanin content and positively affects anthocyanin stability, and color change [17,18].

This spectroscopic method is used to determine quantitative measurements of wavelengths in the visible light region based on the absorption of light by organic chemical compounds in the Ultra Violet (UV) spectrum range, so that this method can measure wavelengths in the photodegradation process which is the process of decomposing organic compounds into simpler compounds with the help of photon energy from UV light [19,20].

APPJ is operated at atmospheric pressure without the use of vacuum pumps, regular maintenance, and expensive space. Current technology has overcome issues such as high breakdown stress, continuous arcing, and instability, thus making APPJ a tool with potential for industrial applications [21,22].

Several types of APP consist of atmospheric Pressure Plasma Jet (APPJ), Dielectric Barrier Discharge (DBD), microplasma, and Corona Discharge (CD) [22]. Various plasma excitation techniques can produce APP with different gas temperatures and charge densities; the synergistic effect of plasma species has high energy and temperature so that it can achieve potential functions in material processing. Atmospheric pressure plasma technology is useful for inversetile applications such as surface modification and thin film deposition. Plasma jets are made up of highly reactive plasma and offer fast processing capabilities. Furthermore, it is a large-area compatible non-vacuum process, which makes it more appealing for DSSC production [23].

This research is focused on natural dye photosensitizers that will be extracted from plants, namely *Medina Lawsonia Inermis, Yemeni Lawsonia Inermis, Caessalpinia Sappan,* and *Areca Catheu*. The selection of these ingredients was because they contain anthocyanins. Natural dye photosensitizers last a lifetime and were analyzed through the photodegradation process. These natural dye photosensitizers are applied in hybrid DSSCs, and their optical, electrical, and electronic properties are characterized, which contribute to their performance. Finally, natural dye photosensitizers will be applied in hybrid DSSCs to determine their conversion ability of light energy to electrical energy.

2. Methodology

2.1 Materials

The main materials involved in this works are naturals dyes extracted from *Medina Lawsonia Inermis, Yemeni Lawsonia Inermis, Caessalpinia Sappan,* and *Areca Catheu*, deionized water, filter paper, and ethanol [24-26]. The materials were purchased from A&T Ingredients Sdn Bhd. The research is divided into several phases, including sample preparation, synthesis of natural dyes, UV-Vis Spectrophotometer testing, FTIR Spectroscopy analysis, and photodegradation evaluation before and after APPJ treatment to determine the dye's stability. The flow of the photodegradation process is shown in Figure 1.

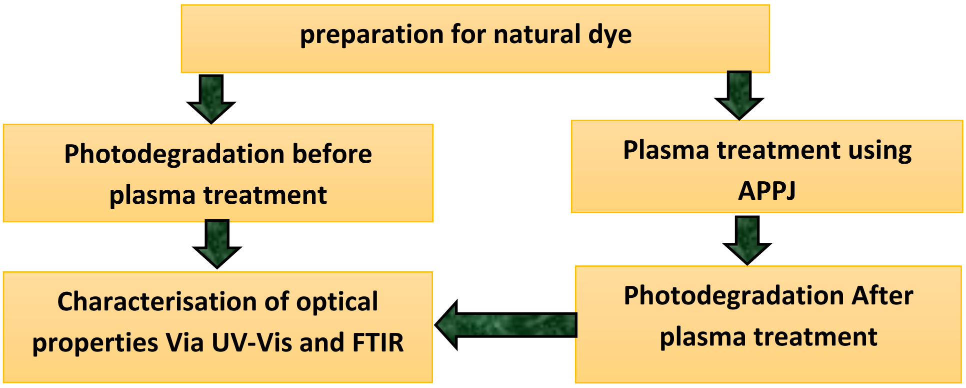


Fig. 1. Photodegradation process of hybrid solar cells

2.2 Extraction of Natural Dye

This stage focusing on sample preparation from *Medina Lawsonia Inermis, Yemeni Lawsonia Inermis, Caessalpinia Sappan,* and *Areca Catheu*. The dye materials are washed thoroughly with deionized water to remove dust and organic impurities before extraction and dried in an oven at 60°C for 12 hours. The dried plants were crushed into fine powder using a grinder (Retsch RM 200). In the natural dye process, 95% ethanol was used in a ratio of 1:10 and then stored in a refrigerator for 3 days. After 3 days, the material was filtered using filter paper (Whatman no.1) [27].

The extracted and prepared dyes were tested using a UV-Vis spectrophotometer to observe the wavelength of anthocyanin and FTIR analysis to identify their molecular composition. The *medina Lawsonia Inermis* and *Yemeni Lawsonia Inermis* extracts produced an intense green color.

Caessalpinia sappan extract produces a deep red colour and *Areca Catheu* produces a red color. The darker the color, the more light and heat it can absorb [27].

2.3 Photodegradation Process

Photodegradation was carried out for 4 weeks directly exposed to sunlight from 9 AM to 4 PM, and data collection was carried out for every week. *Yemeni Lawsonia Inermis* and *Medina Lawsonia Inermis* used a mixture of 1 ml of natural dye extract and 5 ml of ethanol. *Caessalpinia Sappan* and *Areca Catheu* using a mixture of 1 ml of natural dye extract and 10 ml of ethanol. This was because higher extract concentrations can lead to faster degradation rates and the formation of more degradation products. Photodegradation tests were conducted for 4 consecutive weeks.

The photodegradation phase involving natural dyes was conducted to investigate their stability in terms of how long they can maintain their color pigment. The degradation data of the color pigments *Medina Lawsonia Inermis, Yemeni Lawsonia Inermis, Cassalpinia sappan* and *Areca Catheau* wood are presented in percentage form using the formula in Eq. (1).

$$\text{Percentage of photodegradation} = \frac{A_0 - A_1}{A_0} \times 100\% \tag{1}$$

A_0 is defined as the absorbance before exposure, and A_1 is the absorbance after exposure to light.

2.4 Plasma Treatment

This phase involves natural dyes that have been extracted and exposed to APPJ. APPJ uses helium gas with an applied voltage of 10 kV via inverter. Helium gas is supplied from the cylinder gas tank through a gas tube to the gas regulator at 1 L/min [28,29]. Then, the gas travels through the gas cylinder to the inside flow meter system connecting to a quartz glass tube. Plasma treatments were done at different exposure times and in different dye volumes. *Yemeni Lawsonia Inermis* and *Medina Lawsonia Inermis* samples used a mixture of dye extract with 5 ml ethanol. Meanwhile, for *Areca Catheu* and *Caesalpinia Sappan* samples, a mixture of dye extract with 10 ml of ethanol was used. Plasma treatment were conducted under different conditions of treatment times (1, 3, 5 minutes) [17]. The experimental setup of APPJ system is shown in Figure 2.

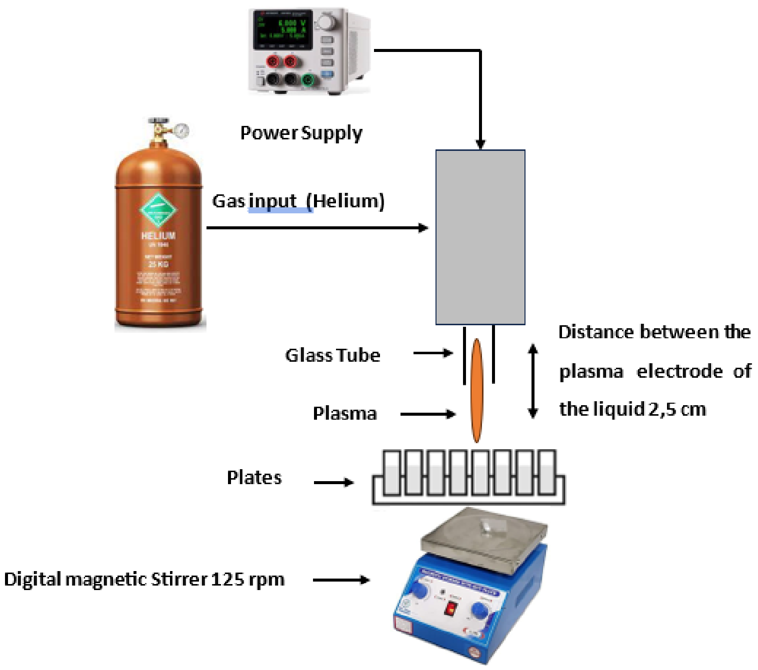


Fig. 2. Schematic diagram of the APPJ system

2.5 Characterisation of Optical Properties

A Ultra Violet-Visible (UV-Vis) spectrophotometer is used to measure the absorbance of samples at specific wavelengths. Light absorption measurements of the natural dye solution were carried out in the wavelength range of 200-900 nm using Perkin Elmer Lambda 25 UV-visible spectrophotometer. The UV-Vis spectrum helps identify the increased absorption of photons in larger-sized particles [30]. For before and after treatment, UV-Vis spectrophotometer test was conducted focusing on the wavelength of anthocyanin.

FTIR characterization of NICOLET IS20 was used to identify compounds and functional groups of anthocyanins in the liquid dye. FTIR analysis can observe the characteristics of natural dyes before and after plasma treatment. FTIR spectroscopy in the spectral range from 500 to 4000 cm^{-1} is used to identify and measure the long-lived infrared active components.

3. Result and Discussion

3.1 Visible Absorption of Natural Dyes Before and After Plasma Treatment

Figure 3-6 show the UV-visible absorption spectrum of the natural dye used in this study. Figure 3 shows that the pigments in *Medina Lawsonia Inermis* absorbed a wide range of visible light from 400 nm to 700 nm. The graph clearly showed that the wavelength of chlorophyll *a* (Chlo *a*) was observed at peaks 414 nm and 665.42 nm, while chlorophyll *b* (Chlo *b*) at peaks 424 nm and 608.04 nm and anthocyanin (Anth) peaks appeared at 468.20 and 535.12 nm. The Figure 4 shows that the absorption spectrum of *Yemeni Lawsonia Inermis* had almost same curve as that of *Medina Lawsonia Inermis*. The differences were maximum absorption at 471.80 nm, which belongs to Anth, and maximum absorption at 664.56 nm, which belongs to Chlo a. Absorption spectrum of *Caessalpinia Sappan* shown in Figure 5 obtained a single broad peak of Anth with a maximum peak at 450.19 nm and a weak peak 545.92 nm. The absorption spectrum of *Areca Catheu* appeared to have broad peaks from 350 nm to 650 nm, as showed in Figure 6 [31,32]. The two broad peaks at 430.91 nm and 465.18 nm showed that beta carotenoid pigment was obtained, while a peak at 578.94 nm belonged to Anth

[33]. The wavelength and intensity of light require organic materials that have colour extracts of a certain colour to absorb more sunlight in photovoltaic devices. Organic materials that are natural materials have fewer side effects or waste than non-organic materials.

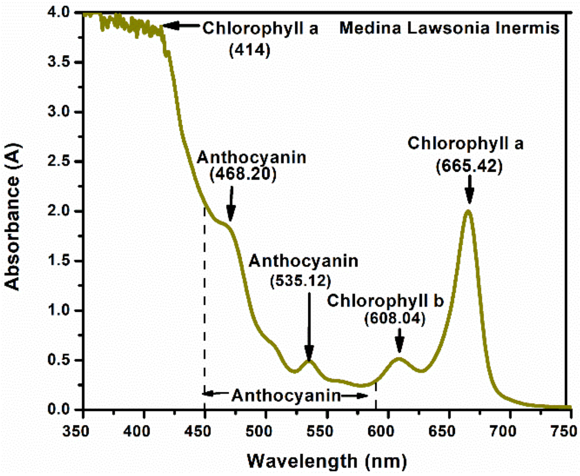


Fig. 3. UV-Vis absorption of *Medina Lawsonia Inermis*

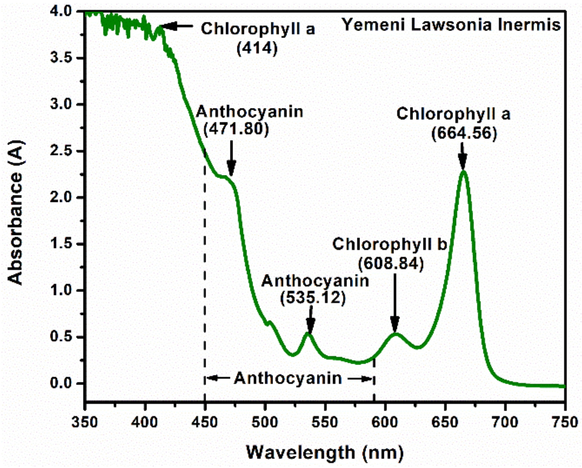


Fig. 4. UV-Vis absorption of *Yemeni Lawsonia Inermis*

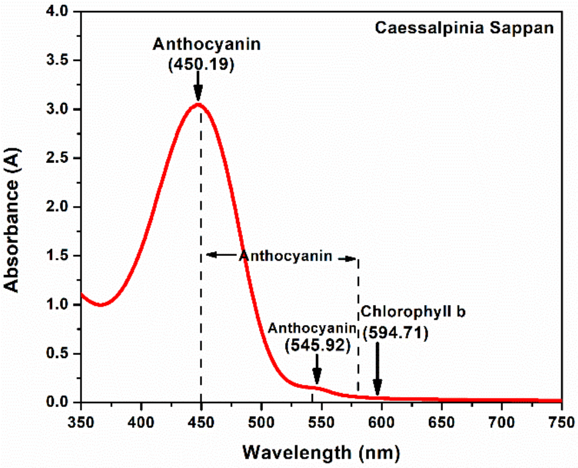


Fig. 5. UV-Vis absorption of *Caessalpinia Sappan*

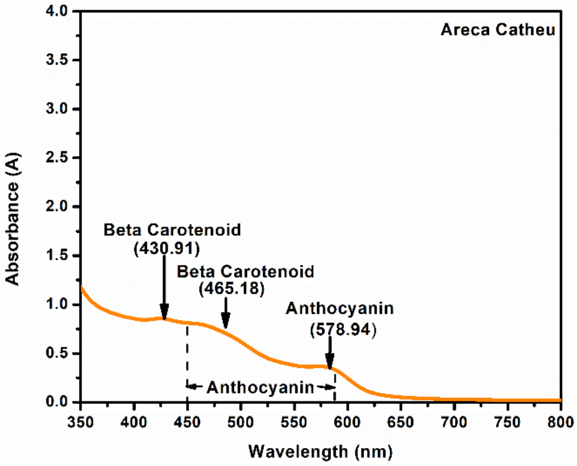


Fig. 6. UV-Vis absorption of *Areca Catheu*

Figure 7-10 showed the UV-visible absorption spectra of natural dyes after APPJ treatment for 1 min, 3 min, and 5 min. *Medina Lawsonia Inermis* dye had anthocyanin peaks at wavelengths of 475.78 nm, 504.60 nm, and 535.96 nm.

Yemeni Lawsonia Inermis had anthocyanin peaks at wavelengths of 476.26 nm, 506.94 nm, and 536.35 nm. *Caessalpinia Sappan* had an anthocyanin peak at a wavelength of 451.74 nm, while *Areca Catheu* had anthocyanin peaks at 579.98 nm. *Medina Lawsonia Inermis*, *Caessalpinia Sappan*, and *Areca Catheu* had the highest absorbance value at 5 minutes of APPJ treatment, while *Yemeni Lawsonia Inermis* at 3 minutes. After APPJ treatment, there was an increase in the anthocyanin wave peak.

The natural colourant also contained chlorophyll, which indicates that the material could absorb sunlight. Anthocyanins' colour and stability are affected by pH, light, temperature, and structure. The colour and stability of anthocyanins are affected by pH, light, temperature, and structure. The pH of 100% ethanol is 7.33, while the pH of pure water is 7.00. The pH of the solution affects the anthocyanin pigment. Anthocyanins are one of the subclasses of phenolic phytochemicals. Anthocyanins are in the form of glycosides (anthocyanidin glycosides and acylated anthocyanins)

[34]. Then again, anthocyanins are the glycosylated form of anthocyanidins. The conjugated bonds of anthocyanins produce red, blue and purple coloured plants [35]. *Medina Lawsonia Inermis* and *Yemeni Lawsonia Inermis* extracts come from the leaves, *Caessalpinia sappan* from the fruit, and *Areca catheu* from the wood. The difference in colour and type of material in the dye will cause different UV-vis absorption differences.

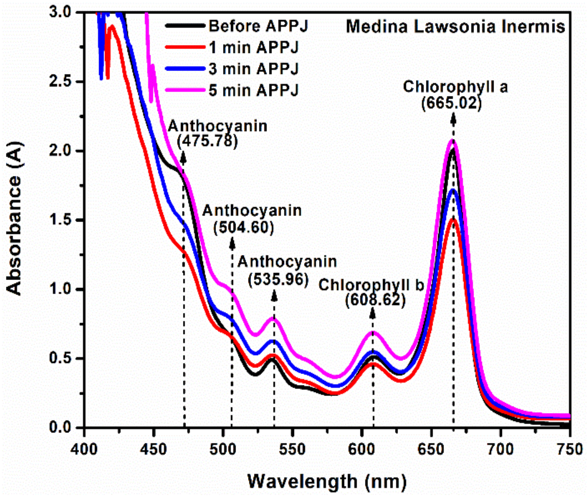


Fig. 7. UV-Vis absorption of *Medina Lawsonia Inermis* after APPJ treatment

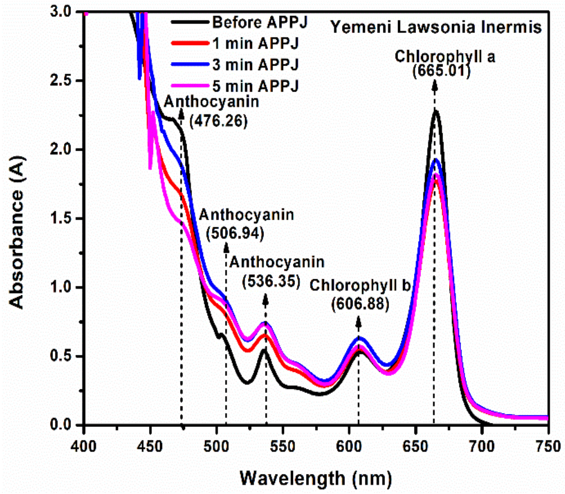


Fig. 8. UV-Vis absorption of *Yemeni Lawsonia Inermis* after APPJ treatment

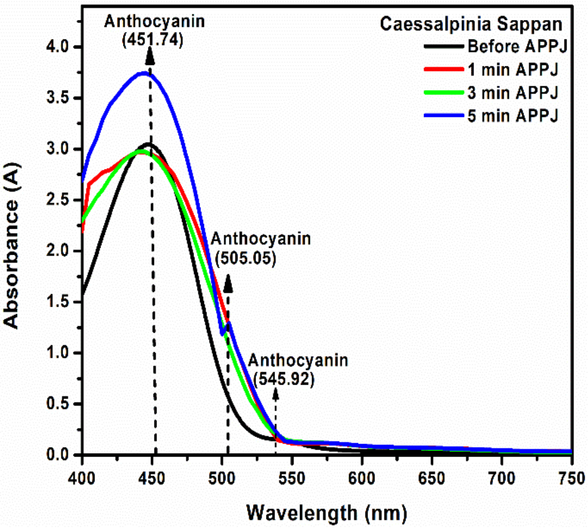


Fig. 9. UV-Vis absorption of *Caessalpinia Sappan* after APPJ treatment

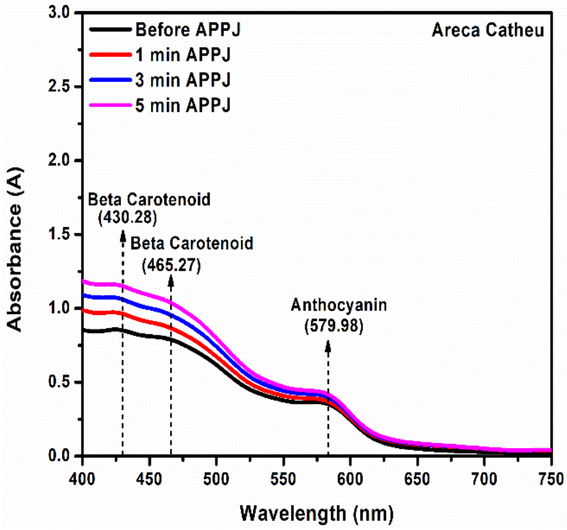


Fig. 10. UV-Vis absorption of *Areca Catheu* after APPJ treatment

3.2 Functional Groups of Natural Dyes

FTIR spectra of natural dye from natural dyes before and after plasma treatment are shown in Figure 11 and 12. Table 1 shows the band value content wavenumber of functional groups of natural dyes before APPJ treatment, while Table 2 shows the wavenumber value content after APPJ treatment. After the FTIR test, in *Medina lawsonia inermis*, *Yemeni Lawsonia Inermis, Caessalpinia Sappan*, and *Areca Catheu* there are frequency peaks of functional groups representing OH, C-H, and C=C functional groups. Hydroxyl (OH) groups are present in anthocyanidins, which are anthocyanins in the form of aglycones, and C=C and C-H groups are present in non-sugar groups bound to anthocyanidins [36,37].

The functional groups found in *Medina Lawsonia Inermis, Yemeni Lawsonia Inermis, Caessalpinia Sappan*, and *Areca Catheu* were -OH, -CH, and C=C. *Medina lawsonia inermis* natural dye before and after plasma treatment has the intensity of OH functional groups (3333.35 cm^{-1} to 3326.35 cm^{-1}), C-H (2970.67 cm^{-1}and 1382.22 cm^{-1} to 2976.12 cm^{-1} and 1382.77 cm^{-1}), C=C (1637.97 cm^{-1} and 879.36 cm^{-1} to 1648.87 cm^{-1} and 878.99 cm^{-1}). *Yemeni Lawsonia Inermis* before and after plasma treatment had intensities of OH (3340.59 cm^{-1} to 3325.93 cm^{-1}), C-H (2870.83 cm^{-1} and 1382.21 cm^{-1} to 2982.38 cm^{-1} and 1384.96 cm^{-1}), C=C (1628.28 cm^{-1} and 879.08 cm^{-1} to 1658.84 cm^{-1} and 878.20 cm^{-1}) functional groups. *Caesalpinia Sappan* before and after plasma treatment has the intensity of OH functional group (3319.59 cm^{-1} to 3330.03 cm^{-1}), C-H (2887.24 cm^{-1}and 1388.02 cm^{-1} to 2972.31 cm^{-1} and 1383.10 cm^{-1}), C=C (1626.05 cm^{-1} and 879.09 cm^{-1} to 1651.76 cm^{-1} and 877.46 cm^{-1}). *Areca Catheu* before and after plasma treatment has the intensity of OH functional group (3326.59 cm^{-1} to 3326.30 cm^{-1}), C-H (2880.24 cm^{-1} and 1388.02 cm^{-1} to 2972.31 cm^{-1} and 1385.34 cm^{-1}), C=C (1637.97 cm^{-1}and 879.35 cm^{-1} to 1651.38 cm^{-1} and 878.20 cm^{-1}).

This observation is parallel to the functional groups found in the molecular structure of Anth, Chlo *a*, Chlo *b,* and beta carotenoid pigments as shown in Figure 13. After these pigments were treated with plasma, it was found that the response of the functional groups was different, where in *Medina Lawsonia Inermis* and *Yemeni Lawsonia Inermis*, the wavenumber of -OH has decreased, while -CH and C=C have increased. The increase and decrease in the value of functional groups before and after plasma treatment are still in the OH, C-H and C=C ranges, this indicates that before and after plasma treatment there is still anthocyanin content in the material. Shows that changes in pigment structure have occurred due to plasma ionization, and plasma-activated compounds can affect changes in pigment structure, breaking down the band value content of natural dye functional groups [38].

The pigment Anth in *Caessalpinia Sappan* showed that after plasma treatment, the wavenumber of -OH and -CH increased, but it only slightly changed at C=C. This demonstrated that -OH and -CH are responsible for the change in Anth pigment molecule structure. While the beta carotenoid pigment in *Areca Catheu* shows that the wavenumber of -OH does not change after plasma treatment, but -CH and C=C increase their wavenumber, meaning that -CH and C=C are ionized by plasma, and there is a structural change in the beta carotenoid molecule.

In Table 1 and Table 2, it can be seen that broad absorption occurs in region *a* with the presence of OH stretching groups and hydrogen bonds as intermolecular interactions between anthocyanins [34]. In regions *b* and *d*, there are C-H stretching groups of alkane compounds that confirm chlorophyll derivatives in natural dyes suitable for UV-VIS absorption. In addition, in region *d*, C-H bending aldehyde compounds were identified. In regions *c* and *f*, C=C stretching alkenes compounds were identified.

The content of band values and functional groups before and after plasma treatment changes in region e, namely before APPJ treatment, identified C-O-C bending anhydride compounds were identified, while after plasma treatment identified CO-O-CO stretching anhydride was identified. This is probably due to the change in oxidation state and the formation of new chemical bonds [39]. In Figure 11, some relatively weaker peaks in the untreated sample now appear stronger after the treatment, as shown in Figure 12.

Although a Helium plasma jet was used, the presence of oxygen under atmospheric conditions may have influenced the changes in functional groups [40,41]. The main difference between different types of anthocyanins is the number of hydroxyl groups and sugar groups bound to the molecular structure or bond position. The spectral data of *Areca Catheu* shows the presence of hydroxyl groups (V_{max} 3400 - 2924 cm^{-1}) [42,43].

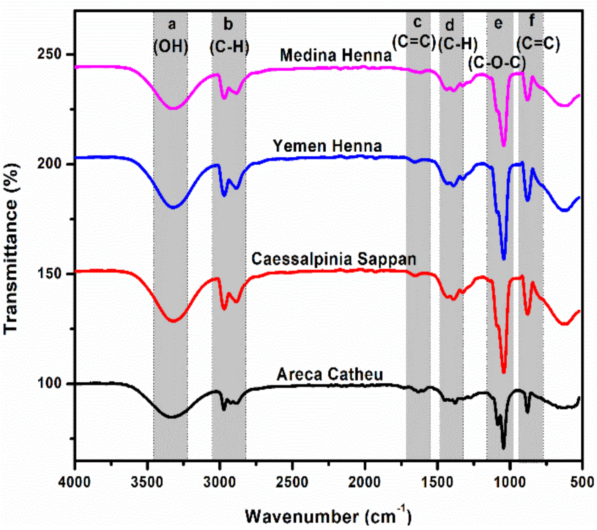


Fig. 11. FTIR spectra of *Medina Lawsonia Inermis, Yemeni Lawsonia Inermis, Caessalpinia Sappan* and *Areca Catheu* before APPJ treatment

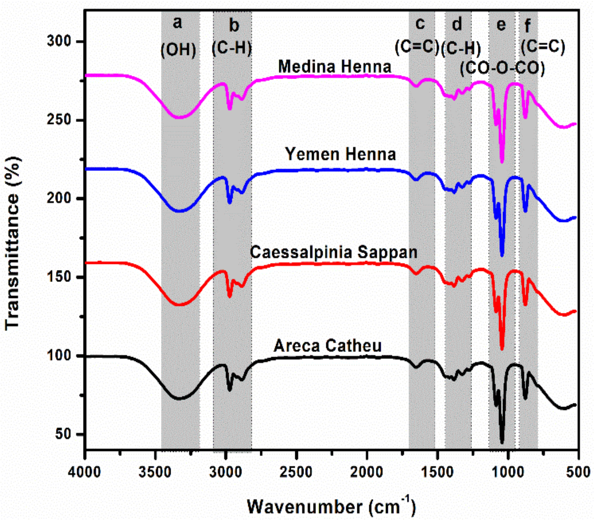


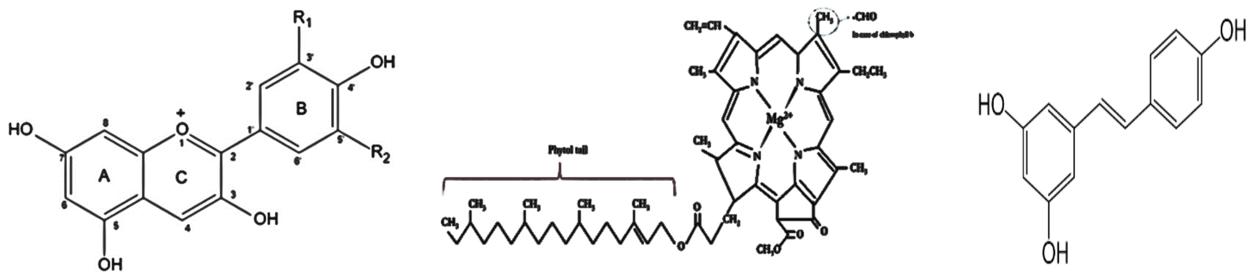
Fig. 12. FTIR spectra of *Medina Lawsonia Inermis, Yemeni Lawsonia Inermis, Caessalpinia Sappan* and *Areca Catheu* after 1 min APPJ treatment

Table 1
FTIR Spectra of natural dyes before plasma treatment

Sample	Intensity (cm^{-1})						Ref.
	a (OH)	b (C-H)	c (C=C)	d (C-H)	e (C-O-C)	f (C=C)	
Medina Lawsonia Inermis	3333.35	2970.67	1637.97	1382.22	1045.31	879.36	[44]
Yemeni Lawsonia Inermis	3340.59	2870.83	1628.28	1382.21	1045.32	879.08	[45]
Caesalpinia Sappan	3319.59	2887.24	1647.66	1388.02	1045.31	879.09	[33]
Areca Catheu	3326.59	2880.24	1626.05	1388.02	1045.31	879.35	[32]

Table 2
FTIR Spectra of natural dyes after plasma treatment

Sample	Intensity (cm^{-1})						Ref.
	a (OH)	b (C-H)	c (C=C)	d (C-H)	e (CO-O-CO)	f (C=C)	
Medina Lawsonia Inermis	3326.35	2976.12	1648.87	1382.77	1041.09	878.99	[46]
Yemeni Lawsonia Inermis	3325.93	2982.38	1658.84	1384.96	1045.88	878.20	
Caesalpinia Sappan	3330.03	2972.31	1651.76	1383.10	1042.15	877.46	
Areca Catheu	3326.30	2972.31	1651.38	1385.34	1042.15	878.20	



(a) (b) (c)

Fig. 13. Molecule structure of (a) Anthocyanin, (b) The chemical structure of typical chlorophyll molecule. Chlo *a* contains methyl group (-CH 3) and Chlo *b* contains aldehydic group (-CHO) instead. (c) beta carotenoid

3.3 Photodegradation of Natural Dyes

3.3.1 Appearance color of solution dye

After 4 weeks before and after APPJ treatment, the dye samples showed discolouration due to degradation. Before APPJ treatment, *Medina Lawsonia Inermis* and *Yemeni Lawsonia Inermis* natural dyes changed color from intense green to colorless and after APPJ treatment from intense green to colorless. *Caessalpinia Sappan* natural dye when before and after treatment changed colour from intense orange to yellow. Meanwhile, *Areca Catheu* color changed from intense red to non-intense red (Figure 14). The natural dyes experienced color degradation after being exposed for 1 week, 2 weeks, 3 weeks, and 4 weeks.

(a) (b)

Fig. 14. Discoloration of (a) Areca Catheu, (b) Caessalpinia Sappan

3.3.2 UV-Visible absorption of photodegraded solution dye

The optical absorption on natural dye solutions that have been photodegraded under sunlight and open areas for 1, 2, 3, and 4 weeks have been shown in Figure 15-22. In this study, the focus has been given to the effect of the chlorophyll and the anthocyanin regions on the DSSCs. As shown in Figure 15-22, the peak of the chlorophyll and anthocyanin waveforms in natural dyes can be seen at various conditions of natural dyes. In natural dyes, there was a decrease in waveform peaks after degradation. The efficiency of degradation in *Medina Lawsonia Inermis* dyes (60.72%), *Yemeni Lawsonia Inermis* (56.51%), *Caessalpinia Sappan* (85.80%), and *Areca Catheu* (62.34%) suggests that exposure to sunlight affected natural dyes. These findings were approved by Escher *et al.,* who found that the color-changing and stability of anthocyanins are influenced by pH, light, temperature, and structure [47]. During photodegradation in DSSC there is a reduction in dye colour intensity caused by various factors such as These factors include chemical degradation of solar cell components, such as ligand exchange of thiocyanate ions with water or hydroxide ions, loss of ligands at high temperatures, and the influence of UV exposure. In addition, the presence of triiodide ions that exchange dye ligands and the sensitivity of specific ligands, such as thiocyanate ions, to degradation plays an important role in dye breakdown in DSSCs [48].

The efficiency of degradation in *Medina Lawsonia Inermis* dyes (60.72%), *Yemeni Lawsonia Inermis* (56.51%), *Caessalpinia Sappan* (85.80%) and *Areca Catheu* (62.34%).

After APPJ treatment, there was an increase in the percentage of degradation efficiency compared to before APPJ treatment. In *Medina Lawsonia Inermis* at 1 minute (71.35%), 3 minutes (70.25%), and 5 minutes (78.09%). Likewise, *Yemeni Lawsonia Inermis* showed an increase in the percentage of degradation efficiency at 1 minute (89.35%), 3 minutes (89.78%), and 5 minutes (78.08%). *Caessalpinia Sappan*, there was a slight decrease in degradation percentage compared to before APPJ treatment, at 1 minute (79.22%), 3 minutes (80.93%), and 5 minutes (77.33%). Thus in *Areca Catheu* at 1 minute (43.45%), 3 minutes (56.32%), and 5 minutes (63.67%) (Figure 23-26).

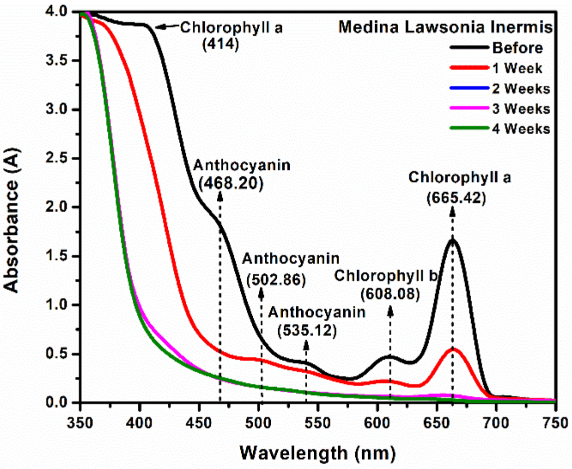


Fig. 15. UV-VIS photodegradation absorption of *Medina Lawsonia Inermis*

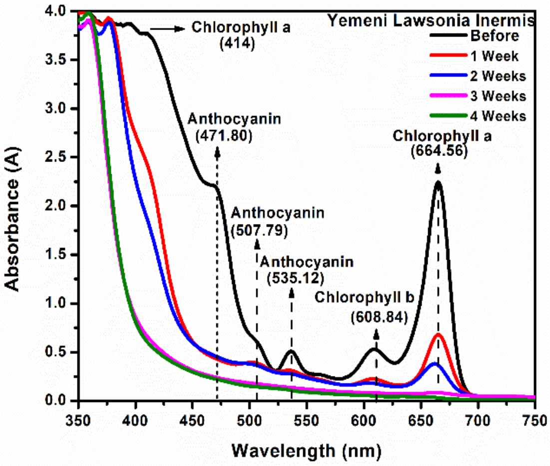


Fig. 16. UV-VIS photodegradation absorption of *Yemeni Lawsonia Inermis*

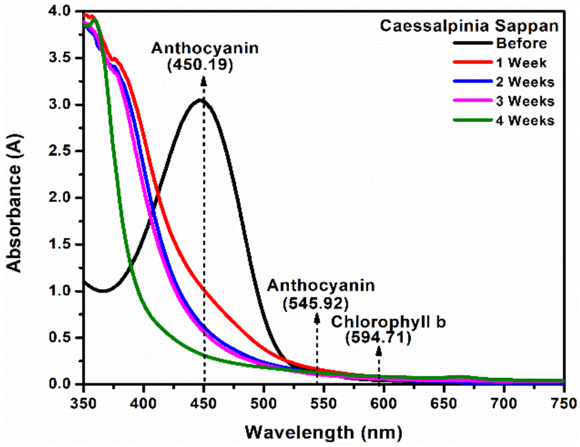


Fig. 17. UV-VIS photodegradation absorption of *Caessalpinia Sappan*

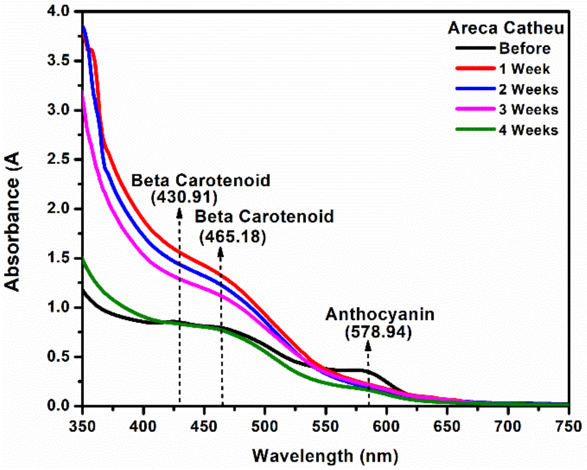


Fig. 18. UV-VIS photodegradation absorption of *Areca Catheu*

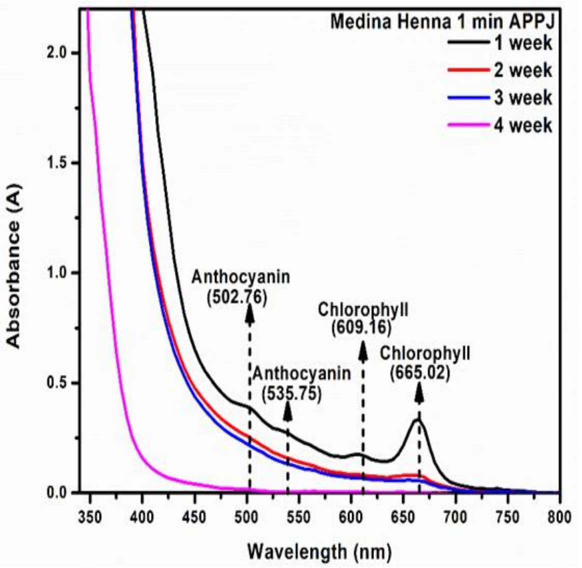


Fig. 19. UV-Visible absorption spectra photodegradation of *Medina Lawsonia inermis* solution that was after 1 minute plasma treatment

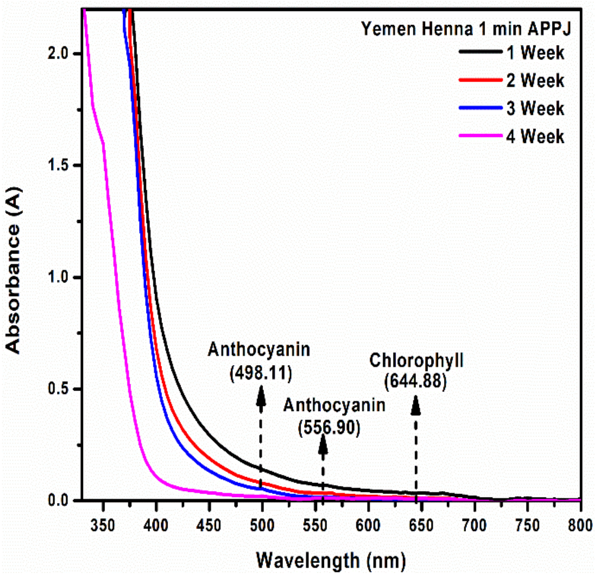


Fig. 20. UV-Visible absorption spectra photodegradation of *Yemeni Lawsonia inermis* solution that was after 1 minute plasma treatment

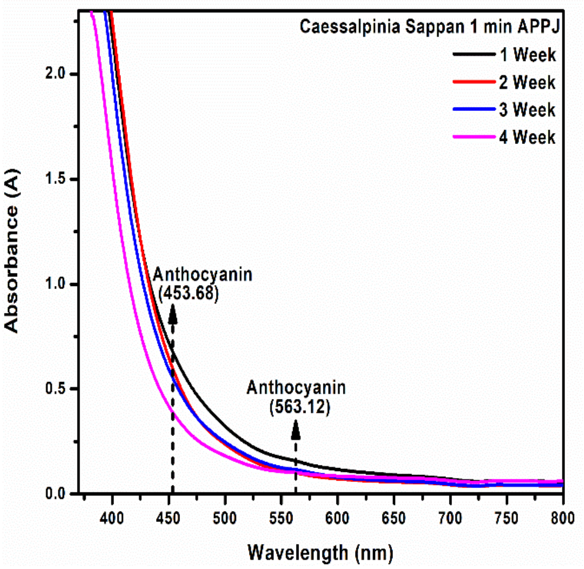


Fig. 21. UV-Visible absorption spectra photodegradation of *Caessalpinia Sappan* solution that was after 1 minute plasma treatment

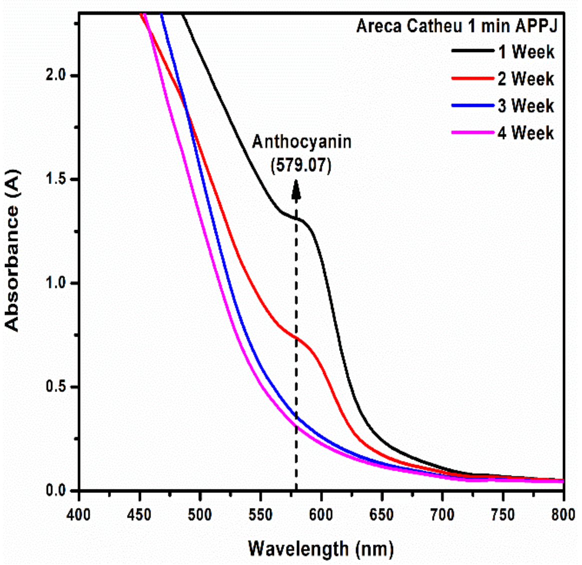


Fig. 22. UV-Visible absorption spectra photodegradation of *Areca Catheu* solution that was after 1 minute plasma treatment

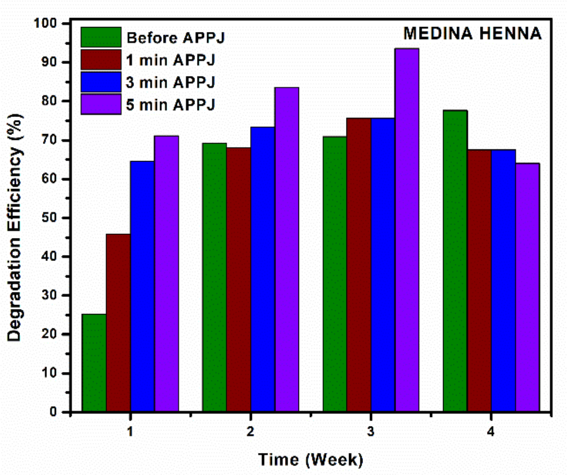


Fig. 23. Degradation efficiency of *Medina Lawsonia Inermis*

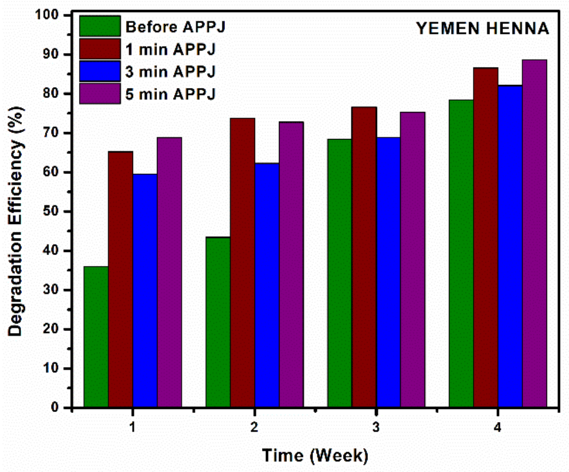


Fig. 24. Degradation efficiency of *Yemeni Lawsonia Inermis*

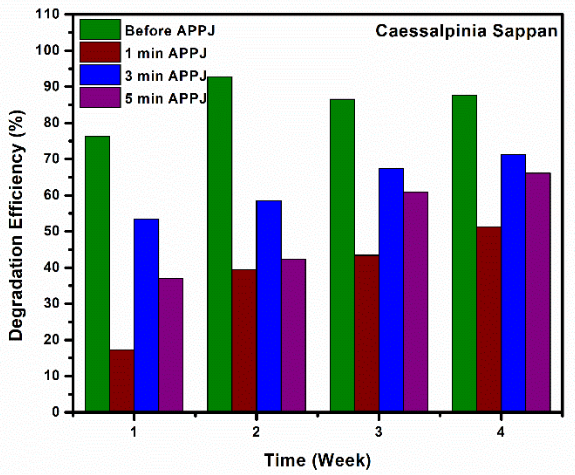


Fig. 25. Degradation efficiency of *Caessalpinia Sappan*

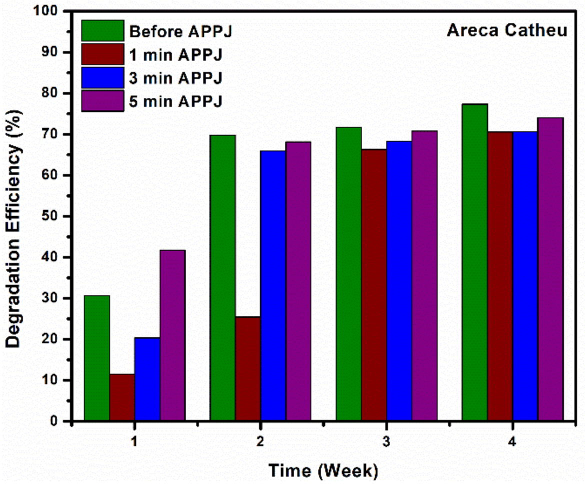


Fig. 26. Degradation efficiency of *Areca Catheu*

3.3.3 FTIR analysis of photodegraded solution dye

The active functional structural groups in anthocyanins and chlorophylls can be seen in Figure 27-30 using FTIR spectroscopy, it would show that the functional groups in anthocyanins and chlorophylls had peak changes due to the photodegradation effect. Details of the wave number changes are recorded in Table 3 and 4.

Table 3 and 4 show the changes in frequency values in the FTIR spectra of extractions before and after APPJ treatment during photodegradation from first week until fourth week. Compounds in natural dyes before and after photodegradation have the same compounds. This shows that the photodegradation process retains the compounds in natural dyes.

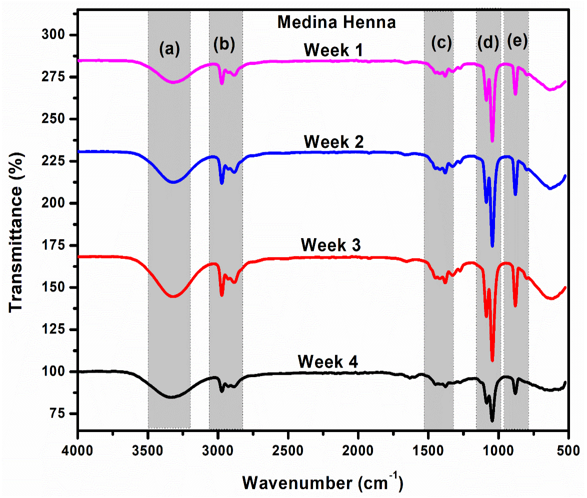


Fig. 27. FTIR spectra of the extraction of *Medina Lawsonia Inermis* after 1 minute plasma treatment

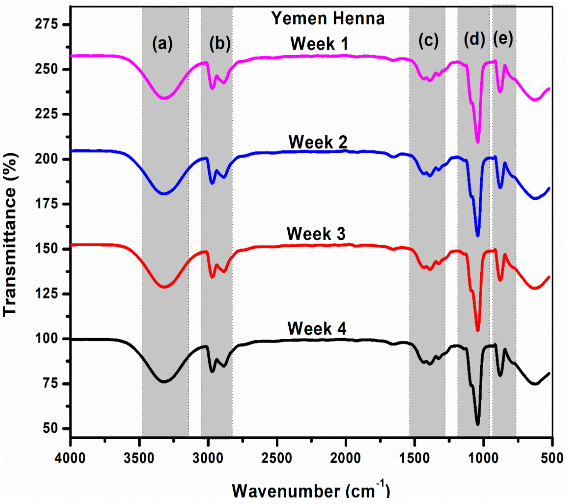


Fig. 28. FTIR spectra of the extraction of *Yemeni Lawsonia Inermis* after 1 minute plasma treatment

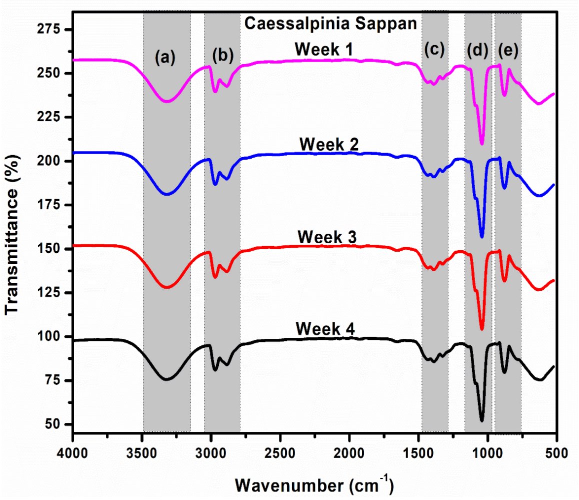


Fig. 29. FTIR spectra of the extraction of *Caessalpinia Sappan* after 1 minute plasma treatment

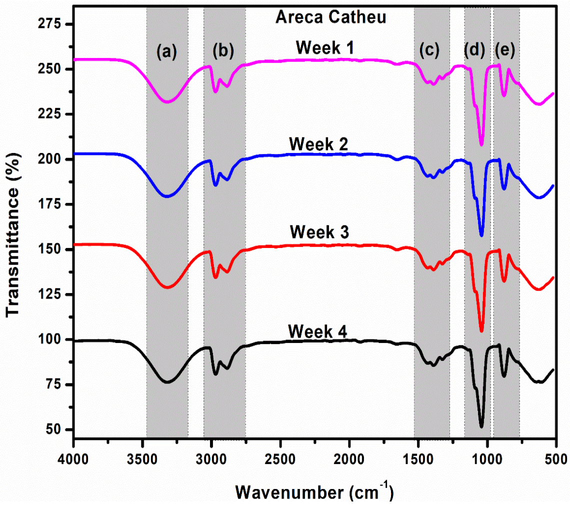


Fig. 30. FTIR spectra of the extraction of *Areca Catheu* after 1 minute plasma treatment

Table 3
The frequency changes in FTIR spectra of the extraction of *the Medina Lawsonia Inermis, Yemeni Lawsonia Inermis, Caessalpinia Sappan* and *Areca Catheu*

Intensity (cm^{-1})	a		b		C		d		e		f	
Period (Week)	(-OH)		(C-H)		(C=C)		(C-H)		(C-O-C)		(C=C)	
	1	4	1	4	1	4	1	4	1	4	1	4
Medina Lawsonia Inermis	3340.15	3343.07	2972.13	2977.44	1638.23	1640.12	1383.23	1384.52	1045.31	1048.21	879.14	879.74
Yemeni Lawsonia Inermis	3340.87	3342.86	2870.93	2968.89	1628.97	1630.13	1382.25	1383.46	1046.13	1047.21	879.08	879.85
Caessalpinia Sappan	3320.19	3323.12	2887.89	2888.14	2887.78	2888.89	1388.47	1389.12	1045.67	1046.15	879.23	879.23
Areca Catheu	3327.12	3329.15	2880.79	2881.03	2881.34	2883.32	1388.23	1389.02	1045.56	1046.13	879.45	879.88

Table 4
The frequency changes in FTIR spectra APPJ treatment of the extraction of the *Medina Lawsonia Inermis,Yemeni Lawsonia Inermis, Caessalpinia Sappan* and *Areca Catheu*

Intensity (cm^{-1})	a		b		C		d		e		f	
Period (Week)	(-OH)		(C-H)		(C=C)		(C-H)		(CO-O-CO)		(C=C)	
	1	4	1	4	1	4	1	4	1	4	1	4
Medina Lawsonia Inermis	3326.75	3327.12	2976.12	2977.13	1649.02	1649.87	1383.05	1383.95	1042.34	1043.16	878.99	878.89
Yemeni Lawsonia Inermis	3325.89	3326.17	2982.12	2982.96	1658.67	1658.90	1384.97	1385.65	1046.13	1046.89	878.56	878.67
Caessalpinia Sappan	3330.04	3330.89	2972.31	2973.11	1652.46	1653.58	1383.56	1385.13	1043.15	1044.23	878.41	878.16
Areca Catheu	3326.30	3326.97	2880.89	2881.18	1652.07	1653.45	1385.69	1385.98	1042.10	1043.02	878.57	878.89

3.3.4 The energy gap of natural dyes before and after degradation

The energy gap value was calculated by extrapolating the linear region of the relationship curve (hv) as abscissa and (ahv) as ordinate until it intersects the energy axis [49]. Table 5 showed that *Medina Lawsonia Inermis* and *Yemeni Lawsonia Inermis* after exposure to sunlight for 4 weeks, had an increasingly large energy band gap value and this indicates that they experience a wider energy gap. Whereas in *Caessalpinia Sappan* and *Areca Catheu*, the energy band gap value is getting smaller, and this indicates that the energy gap is narrower.

Table 6-8 show the energy gap value after APPJ treatment. The energy gap value decreased after photodegradation for 4 weeks in *Medina Lawsonia Inermis* and *Areca Catheu* with 1,3 and 5 minutes of APPJ treatment, while in *Caessalpinia Sappan* there was a decrease in energy gap value at 3 minutes of APPJ treatment. *Yeman Lawsonia Inermis* experienced a significant increase in energy gap value. The magnitude of the band gap energy is influenced by various factors such as temperature. The higher the temperature, the band gap energy tends to be more significant**.**

Table 6-8 show that After APPJ treatment before photodegradation, *Medina Lawsonia Inermis* had a percentage decrease in energy gap at 1 minute (13.91%), 3 minutes (6.43%), and 5 minutes (4.41%). *Yemeni Lawsonia Inermis* had a percentage decrease in energy gap after treatment of 1 minute (3.21%) and 3 minutes (1.02%). *Caessalpinia Sappan* had a percentage decrease in energy gap after 1 minute (39.04%), 3 minutes (37.98%), and 5 minutes (42.96%) treatment. *Areca Catheu* percentage decreased the energy gap in the treatment of 1 minute (8.70%), 3 minutes (9.51%), and 5 minutes (3.19%).

Table 5
Optical Energy Gap photodegradation before APPJ treatment

Natural Dye Sample	Optical Energy Gap (eV)		
	Before expose sun light	After 2 Week expose sun light	After 4 Week expose sun light
Medina Lawsonia Inermis	2.766	3.207	3.172
Yemeni Lawsonia Inermis	2.641	3.052	3.230
Caessalpinia Sappan	3.957	3.226	3.022
Areca Catheu	3.847	3.296	3.701

Table 6
Energy Gap photodegradation after 1 minute APPJ treatment

Natural Dye Sample	Optical Energy Gap (eV)		
	Before expose sun light	After 2 Week expose sun light	After 4 Week expose sun light
Medina Lawsonia Inermis	2.381	1.959	1.763
Yemeni Lawsonia Inermis	2.556	3.122	3.386
Caessalpinia Sappan	2.412	2.894	2.996
Areca Catheu	3.512	2.389	2.369

Table 7
Energy Gap photodegradation after 3 minutes APPJ treatment

Natural Dye Sample	Optical Energy Gap (eV)		
	Before expose sun light	After 2 Week expose sun light	After 4 Week expose sun light
Medina Lawsonia Inermis	2.588	2.337	2.095
Yemeni Lawsonia Inermis	2.614	3.113	3.342
Caessalpinia Sappan	2.454	2.409	2.389
Areca Catheu	3.481	2.376	2.374

Table 8
Energy Gap photodegradation after 5 minutes APPJ treatment

Natural Dye Sample	Optical Energy Gap (eV)		
	Before expose sun light	After 2 Week expose sun light	After 4 Week expose sun light
Medina Lawsonia Inermis	2.644	1.833	2.461
Yemeni Lawsonia Inermis	2.732	3.120	3.348
Caessalpinia Sappan	2.257	2.676	2.796
Areca Catheu	3.724	2.363	2.433

4. Conclusion

This study examined the UV absorption spectra of natural dyes, focusing on the anthocyanin content. The presence of chlorophyll in natural dyes indicates that the material can absorb sunlight. The colour of natural dyes can be seen through changes in absorbance spectra (UV-vis analysis) and shifts in functional group peaks (FTIR analysis) before and after APPJ treatment.

Photodegradation was carried out for 4 weeks with direct exposure to sunlight, showing discolouration due to degradation. After APPJ treatment, the percentage of degradation efficiency increased further and strongly in *Medina Lawsonia Inermis and Yemeni Lawsonia Inermis* while *Caessalpinia Sappan* and *Areca catheu* showed little change in the percentage of degradation compared to before APPJ treatment. Natural dyes have different energy band gap values before and after APPJ treatment, which indicates that the wider and smaller energy gaps are affected by temperature. APPJ treatment is very good for *Inermis Lawsonia Medina, Inermis Lawsonia Yaman, Caessalpinia Sappan* and *Areca Catheu* with treatment for 1, 3 and 5 minutes because the energy gap level decreases, making it easier for electrons to move from the valence band to the conduction band. After APPJ treatment, *Caessalpinia Sappan* had the highest presentation value of energy gap reduction. APPJ treatment has a strong and good effect on natural dyes for degradation efficiency and energy gap level. More experiments are needed to explore this field to improve the performance of natural dyes using APPJ treatment for solar cell applications that are cost-effective, environmentally friendly and have good durability.

Declaration of Conflict Interest

The authors declare that they have no known conflict interests or personal relationships that could have appeared to influence the work reported in this paper.

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