Conductivity Study of Chlorophyll: Potential Solar Panel

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Abstract— **Aim of this research is to investigate the ability of the chlorophyll extracted from oil palm trees, *Elaeis Guineensis*, to conduct electricity in order to replace the conventional dye used in current dye-sensitized solar cells (DSSC). This is because the energy conversion performance of natural dye sensitized solar cells (DSSC) mostly relies on sensitizer. The chlorophyll was extracted using 96% ethanol and was dried in an oven at 80°C. About 2 g of chlorophyll were obtained after drying. The functional anchoring groups of the *E. guineensis* chlorophyll were analysed using Fourier transform infrared spectroscopy (FTIR) and it was found that there were eight functional groups. 10 mg of dried chlorophyll was mixed with deionized water and 10 mg of carboxymethylcellulose powder which were then pressed to form 2 cm by 2 cm size of chlorophyll gel. Lastly, the chlorophyll was measured its conductivity using a multimeter. The amperage measured from the chlorophyll gel is 0.00018A which indicates that the chlorophyll gel is capable to conduct an electricity, hence has the potential to substitute the synthetic dye sensitizer in DSSC.**

Keywords— **Chlorophyll, dye sensitizer, solar cells**

I. Introduction

The fossil fuels have become the source that provide the generation of today's electricity and fulfill approximately 60% of the current total world energy demand. This dependence of nonrenewable fossil fuels has put the world in a precarious situation. Besides, the increasing demand on fossil fuels consumption will lead to depletion of natural resources and soon the fossil fuels will become limited. The uses of today's non-renewable energy resources such as coal also contributes the carbon byproducts which promotes the buildup of air pollution, global warming and ultimately greenhouse effect. Since energy is an essential requirement for human survival, it is therefore vital to find alternative ways to produce electricity in a clean, efficient and sustainable manner.

The Earth receives energy from the sun at rate of approximately 12 × 1017 J s-1 which has exceeded the yearly worldwide energy consumption rate at approximately 1.5 × 1013 J s^{-1}[14]. This may offer a wide opportunity to devise an approach for the effective capture and storage of solar energy as the alternative way in replacing the non-renewable sources. In 1991, dye-sensitized solar cells (DSSC) technology was first discovered by Michael Gratzel and Brian O'Regan [6]. DSSC technology is an economical source of renewable energy that converts solar energy into electrical energy which is very dependent on the material and the quality of semiconductor electrode and the dye sensitized used to ensure the effective performance. There are four components in DSSC which are photoanode, dye-sensitizer, electrolytes, and counter electrode.

Obviously, the source of light for plants to undergo the process of photosynthesis is sunlight in the form of solar energy. The tiny particles that travel as lights stream is called photons. The energy carried by the discrete particles is known as quantum and it is related to its wavelength. Theoretically, the shorter the wavelength, the greater the energy contained in its quantum [4]. In green leaves, there are two groups of photosynthetic pigments which are Pigment System I (PS I) that is associated with both cyclic and non-cyclic phosphorylation and Pigment System II (PS II) that is associated with only non-cyclic phosphorylation. Both of the pigments each consists of a central core complex and light harvesting complex. The core complex is associated with electron donors and acceptors. PS I which is found in thylakoid membrane and stroma lamella contains pigments chlorophyll-a 660, chlorophyll-a 670, chlorophyll-a 680, chlorophyll-a 690 and chlorophyll-a 700 where the chlorophyll-a 700 (P_{700}) is the reaction centre for PS I. On the other hand, for PS II, it is found in thylakoid membrane only and it contains chlorophyll-b 650, chlorophyll-a 660, chlorophyll-a 670, chlorophyll-a 678, chlorophyll-a 680-690 and also phycobillins. [4]

There are two phases involved in photosynthesis which are light reaction and dark reaction. In light reaction, the reaction is completely dependent upon the light where it takes place in thylakoid membrane. The three stages involved in light reaction are excitation of chlorophyll, photolysis of water and phosphorylation. During excitation of chlorophyll, the P_{680} or P_{700} of PS I and PS II are stroked by photons, they receives quantum of light and become excited, thus releasing electrons. The equation involved at this stage is as below:

$$Chlorophyll \rightarrow CHl^{+}(oxidised) + e^{-}(electron)$$

For the photolysis of water which is the splitting of water molecules, Van Neil verified that the source of oxygen evolved in the reaction is from water molecules, H_2O, not from carbon dioxide [16]. Two molecules of water are required during photosynthesis in order to release on molecule of oxygen. The equations involved in photolysis of water are as below:

$$4H_2O \rightarrow 4H^+ + 4(OH^-)$$
$$4(OH) \rightarrow 4(OH) + 4e^-$$
$$4(OH) \rightarrow 2H_2O + O_2 \uparrow$$
$$2H_2O \rightarrow 4H^+ + 4e^- + O_2 \uparrow$$

(J. Franck, 1964)

The synthesis of ATP from ADP and inorganic phosphate in presence of light is called photophosphorylation. There are two types of photophosphorylation which are cyclic and non-cyclic. For cyclic photophosphorylation, the excited PS I expel electron and passing through a series of electron carriers including FeS, ferredoxin, plastoquinone, cytochrome b-f complex and plastocyanin where the PS I is then become oxidized. The expelled electron is then returned to PS I. However, during the electron passing through the ferredoxin and plastoquinone, the electron loses sufficient energy to form ATP from ADP and inorganic phosphate. [4]

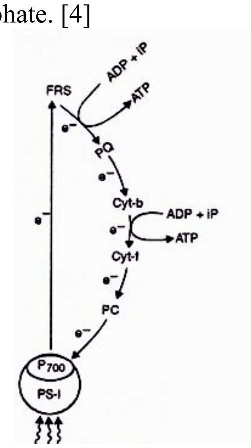

Fig.1: Cyclic Photophosphorylation [13]

In contrast, the non-cyclic phosphorylation, the electron expelled by the PS I and PS II does not return as in cyclic phosphorylation. The reaction of PS II is at P_{680} where the electron is then released during the photolysis of water that takes place on the inner side of the thylakoid membrane, passing through the electron carriers such as quinone (Q), plastoquinone (PQ), cytochrome b-f complex and lastly plastocyanin before the electron is ready to be picked up by the PS I which the reaction centre is at P_{700}. Nevertheless, the electron loses sufficient energy while passing over cytochrome complex to form ATP. Again, as the PS I absorbs light, the electron is extruded from the reaction centre, P_{700}. The electron is handed over to the Ferrodoxin Reducing System (FRS), ferrodoxin (Fd) and NADP reductase where the $NADP^+$ is reduced to $NADPH_2$ through the releasing of H^+. The H^+ ions that are released by the electrons are pumped to the thylakoid membrane to create a proton gradient at which the gradient will trigger the coupling factor to synthesize ATP.

During the transportation of electrons through the photosystems, there are protons that are transported across the thylakoid membrane due to the primary acceptor of electron which is located towards the outer side of membrane that transfer its electron to the H carrier. While transporting the electron through the electron carriers, the molecule removes a proton from the stroma, releasing into the inner side of lumen membrane. As the electron arrives at the NADP reductase enzyme which is located on the stroma, proton is essential for the reduction of $NADP^+$ to $NADPH_2$. Hence, the proton released are then removed from the stroma. As the result, the concentration of proton in the stroma decreases while the concentration of proton in the lumen increases, creating a proton gradient.

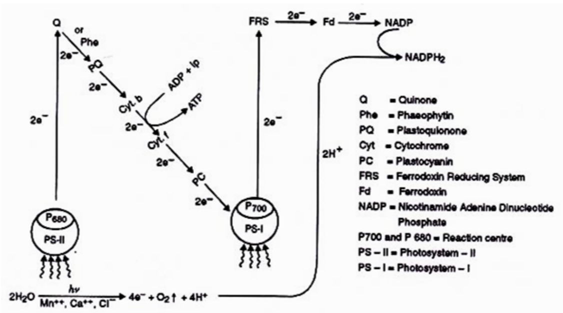

Fig. 2: Non-cyclic Phosphorylation [13]

From the Figure 2, the light reaction produces three types of products which are essential for the nature. For instance, oxygen molecules are produced from the process of photolysis of water, ATP is produced during the phosphorylation and $NADPH_2$ is produced from the reduction reaction at the NADP reductase enzyme. The oxygen molecules are then to be diffused out of the chloroplast whereas ATP and $NADPH_2$ are used for the biosynthetic phase of photosynthesis to synthesis sugar. The biosynthetic phase occurs during the absence of light.

There are two categories of dye sensitizer which are synthetic dye and natural dye. At the meantime, the most effective DSSC is the one that use complex compound Ruthenium (II) polypyridyl with an overall efficiency of 10% [2]. Meanwhile, the natural dye used as dye sensitizer is from natural sources such as chlorophyll which is extracted from plant leaves. The simple and low operating cost procedure to extract the chlorophyll have trigger the researchers to develop natural dye-sensitized solar cells that are more environmental friendly. However, the natural dye has the disadvantage of being easily degraded at high temperature and less efficient.

Titanium oxide, TiO_2 which is extensively used as the semiconductor material in DSSC has a mesoporous structure. It is chosen as the material in DSSC due to its advantages of being inert, stable against corrosion, non-toxic and possess a large bandgap. The performance of DSSC depends on the number of dye molecules that can be absorbed by the TiO_2 layer. The molarity of adsorbed dye solution when dipping the TiO_2 layers in dye solution influence the number of dye molecules adsorbed on the TiO_2 layer [16]. The chlorophyll dye used as the photosensitizer mimic the process of photosynthesis in green plants. Chlorophyll produces strong absorption wavelength 420 nm and 660 nm [10] The higher the absorption peak of a dye, the greater the photon energy can be absorbed and converted into electrical energy in the solar cells application.

According to Gratzel, there are few characteristics of an efficient dye sensitizer must portray which are able to absorb light in the visible region, good attachment on the surface of photoelectrode for fast electron transfer, the excited state of dye must be slightly above the TiO_2 conduction band and its ground state level below the redox potential of the electrolyte, and lastly the dye must easily accepting replacement electron from electrolyte. The chlorophyll will be extracted using the conventional method which is by soaking in organic solvent. It is reported in Journal of the World Aquaculture Society that chloroform has high efficiency in extracting chlorophyll-a compared to 90% acetone and absolute methanol. Nevertheless, due to its toxicity and it high volatility, chloroform is not popular to be used for extracting chlorophyll.

Basically, there are three major processes that take part in the principle of dye sensitized solar cells (DSSC) which are photon strike, electron excitation and electricity generation. This can be seen that the working principle of DSSC is similar to the mechanism of photosynthesis in plants. Generally, the layer of dye which is placed at the surface of a semiconductor absorb the light emitted from the light source. The electron generated by the dye (D) molecule undergo an excited state, thus has the ability to expel electron into the conduction band of the semiconductor. The electron is transported to the cathode which act as the electron collecting counter electrode.

Below are the equation involved in the main process of DSSC:

$$D + hv \rightarrow D^*$$

The dye (D) molecules absorb photon from the light (hv) emitted and produce excited state dye (D^*) molecules that lies above the edge of conduction band TiO_2 that will oxidized to D^+ [12].

$$D^* + TiO_2 \rightarrow D^+ + e^-(TiO_2)$$

The excited state dye (D^*) molecules injects an electron to the TiO_2 conduction band and the deactivation reaction occurs to relax the excited state of dye (D^*) molecules. The deactivation reaction occurs simultaneously with the injection of electron.

$$D^* \rightarrow D$$

The oxidation of iodide and reduction of iodine in the electrolyte occurs as below where the oxidized dye (D^+) accept electrons from the iodide ion (I^-):

$$2D^+ + 3I^- \rightarrow 2D + I_3^-$$
$$I_3^- + 2e^-(catalyst) \rightarrow 3I^-$$

The triiodide ions (I_3^-) is reduced and the iodide ion is now restored, thus completing the circuit and back to original state ready for next cycle.

In the external circuit, the current flow is present due to the injection of electron and hence provide electron for the reduction of iodine at the electrolyte to occur. The collection efficiency of the photo-injected electrons at the anode back contact is hindered by the processes of back electron transfer and TiO_2 conduction band electron capture by redox reaction [5]. The equation for the recombination processes are as below:

$$D^+ + e^-(TiO_2) \rightarrow +TiO_2$$
$$I_3^- + 2e^-(TiO_2) \rightarrow +3I^- + TiO_2$$

From the processes and equation explained above, it can be conclude that the working principle of dye-sensitized solar cells (DSSC) employ the process of photosynthesis. From the Fig. 3, it can be seen that the dye molecules act as reaction centre P_{680} at PS II. The iodide act as water and the anode behavior is similar to the electrons transferred to reaction centre P_{700} at PS I where in DSSC system, excited electron from the dye molecules is transferred to be countered at the cathode, while in photosynthesis, excited electron is transferred passing through electron carrier from P_{680} to P_{700}.

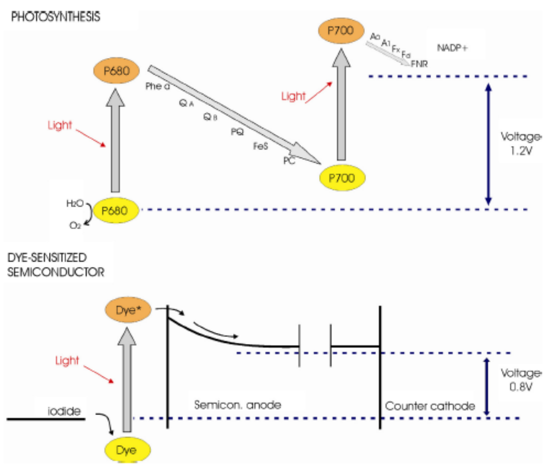

Fig. 3: Comparison between Photosynthesis and DSSC system [5]

There are various studies have been made on the types of dye from plants to substitute ruthenium complex dyes. As for now, dye molecules from ruthenium complex possess the highest efficiency of 11-12% [9]. Nevertheless, it is reported that ruthenium complex as dye sensitizer is non environmental friendly, expensive and difficult to fabricate. Plus, it is also toxic since it has heavy metals.

Several natural plant-based dyes have been established to study their efficiencies as sensitizers in DSSC. For example, spinach leaves were extracted its chlorophyll using two different solvents which are ethanol and deionized water. The performance of DSSC using chlorophyll from spinach leaves as dye was also measured using solar simulator 100 mW/m^2 Ketley 2450. The result obtained shows that the spinach chlorophyll extracted using DI water has higher fill factor (FF) with 0.49% compared to using ethanol as solvent with 0.36%. This may due to the broad absorption wavelength [18].

Moreover, there are also study of natural plant-based dyes using Aloe Vera and Cladode as sources. The Aloe Vera displayed an absorption peak of 524 nm while Cladodes has 662 nm absorption peak. From the result obtained from the study, Cladode as dye sensitizer that placed on the TiO_2 film on FTO conductive glass has highest conversion efficiency at 0.74% compared to Aloe Vera at 0.38% [8].

In the study by Giuseppe [5] brown seaweed pigments were used as to obtain the chlorophyll. Due to the important characteristics of dye sensitizer must portray which is to have strong binding to TiO_2 film, the dye sensitizer must have carboxylic groups for anchoring. Since chlorophyll-a has weak interaction of ester and ketocarbonyl groups with hydrophilic oxide surface, dye sensitizer with chlorophyll-a does not adsorb on TiO_2 efficiently [2]. It is found that chlorophyll-c which is most abundantly belongs to seaweeds and marine plankton have the terminal carboxyl group which allows efficient of anchor of dye molecules to mesoporous TiO_2 [19].

Brown seaweeds, *U. pinnatifida* were dried and minced. They were extracted their chlrophyll by soaking in a solution of 90% acetone: 10% DI water. The dye solution was prepared to deposit onto TiO_2 film as dye sensitizer. From the photoelectrochemical studies, the performance of DSSC using brown seaweed as dye sensitizer has an efficiency of 0.178% [5].

II. METHODOLOGY

The oil palm tree, *Elaeis guineensis* which is planted around Tasik Seksyen 7 in Shah Alam, Selangor was to be collected its leaves and all the leaves' veins were cut off. The collected leaves were cut into small pieces in order to have large total surface area for extraction later on. The small pieces of leaves were weighed on a weighing balance for 250 gram. They were placed in a 500 mL beaker.

A. Chlorophyll Extraction

The leaves are fully immersed with 200 mL of 96% of ethanol and there were transferred into a grinder. The leaves were ground for 5 minutes. After 5 minutes, the clear solution initially has turned to dark green. The extracted chlorophylls were filtered out and were poured into clean 250 mL beaker. The extracted chlorophylls were placed in a dark place for 48 hours. The chlorophylls were to be dried by placing them in a Memmert Oven at 80°C in order to obtain the dry mass. The dry mass chlorophylls were weighed using electronic balance.

B. Characterization of chlorophyll using Fourier transform infra-red spectrometry (FTIR)

The presence of any changes in functional groups of the chlorophyll was measured by FT-IR Spectroscopy. Approximately 2 mg of dried chlorophyll were pressed at the transparent pellet. Spectra were viewed using Perkin Elmer Spectrum One machine. The spectra produced are transmittance mode between wave numbers of 4000 cm-1 and 500cm-1**.**

C. Preparation of chlorophyll gel

Approximately 10 mg of chlorophyll were added with deionized water to make chlorophyll solution and were mixed with 10 mg of carboxymethylcellulose. The chlorophyll gel was pressed between two glass lids into a square size of 2 cm x 2 cm to ensure the chlorophyll is in stable form.

D. Conductivity of chlorophyll using multimeter

The 2 cm x 2 cm chlorophyll gel was connected to positive and negative terminals of Sunwa YX-360TRD multimeter.

III. RESULTS AND DISCUSSION

The result of the chlorophyll extracted after drying weighed about 2 g of dry mass. Figure 4(a) shows the chlorophyll extracted using ethanol and Figure 4(b) shows the chlorophyll after drying at 80°C.

Fig. 4(a): Chlorophyll extracted with ethanol

Fig. 4(b): Chlorophyll after drying at 80°C

The use of FTIR fingerprinting for chlorophyll extract tends to focus on identification and assessment of the stability of the chemical constituents functional groups observed by FTIR analysis. The results of FTIR fingerprint for the oil palm leaves extracted using ethanol is shown in Fig 5. The results of functional group analysis using FTIR had demonstrated the existence of various functional groups in the *E. guineensis* extract. This was verified by the peaks formed during the FTIR study (Fig. 5). There were eight major peaks in the range of 1000 – 1700 cm^{-1} and 2800 – 3300 cm^{-1} observed in the FTIR spectra.

The FTIR spectrum of *E. guineensis* presents a stretching absorption band and bending vibration at 3272.88 cm^{-1} which relies in between the broad shape that occurs at range 3600-2600 cm^{-1} elucidate the O-H stretch. Within the same range, the peaks present at 2917.52 cm^{-1} and 2849.53 cm^{-1} are assigned to aliphatic hydrocarbon group $-CH_2$ and $-CH_3$ respectively. The peak at 1617.25 cm^{-1} reveals the presence of C=O stretch. Thus the presence of carbonyl group in the chlorophyll is observed through the O-H stretch and C=O stretch. There is also another peak at 1036.6 cm^{-1} that corresponded to the vibration of the O-H associated bond of water molecules which are hydrogen bonded hydroxyl groups of cellulose and absorbed water [1]. The peaks present at 1239.55 cm^{-1} and 1357.57 cm^{-1} in extracted *E. guineensis* chlorophyll are assigned to C-C-H bending and C-C stretching vibrations respectively [3].

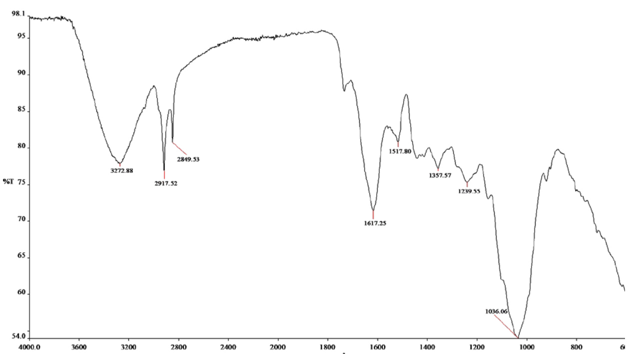

Fig 5: FTIR spectra of E. guineensis chlorophyll

The determination of the amperage of the chlorophyll was obtained by measuring using multimeter. The amperage value produced was 0.0018 A. The preparation for the amperage measurement was as in Fig 6.

Fig. 6: 2cm x 2cm Chlorophyll gel connected to multimeter

The value of the amperage obtained indicate that the chlorophyll is able to conduct an electricity since there are movement of electrons within the chlorophyll. The light in term of photons strike was to be absorbed by the chlorophyll molecules. The electron generated by the chlorophyll molecules undergo an excitation state, thus has the ability to expel electrons.

In dye sensitized solar cell (DSSC), the most vital component that determines and ensures the efficiency of the solar cell is the sensitizer. Dye chlorophyll plays a big role in absorbing light for energy harvesting process as well as the electron excitation process to enable the electron injection from dye to the semiconductor. Since DSSC employs the process of photosynthesis, the dye sensitizer acts as the reaction centre P_{680} at PS II.

The addition of solvent such as 96% of ethanol for the extraction purpose was reported by Rodhi et al, 2018 [16] that it can improve HOMO - LUMO level energy which can upsurge the ability of electron injection and also reduce bandgap energy. The distance between the valence band of electrons and the conduction band is known as the band gap at which it represents the minimum required energy to excite an electron to a state where it can later participate in conduction [11].

For semiconductors, the band gap is quite small enough that some excitation of electrons produced due to the photon striking can bridge the conduction band, thus conducting some electricity. For the case of DSSC, it is proven that the dye molecules from the chlorophyll play the vital role in capturing the photons strike by the Sun energy to act as photosensitizer, thus allowing the excitation electrons reach the conduction band of the semiconductor.

IV. CONCLUSION

In this study, oil palm tree leaves were employed to measure its ability to conduct electricity in order to replace the conventional dye as the photosensitizer in the DSSC. The characterization of chlorophyll extracted from *Elaeis Guineensis* using ethanol in the FTIR spectrum showed eight major types of functional groups available in the sample. The chlorophyll was prepared into a solid form of chlorophyll gel to ensure its stability. The amperage value of the chlorophyll obtained from the multimeter which is 0.0018A indicate the conductivity of the chlorophyll due to the electrons excitation generated. Since the photocurrent response of DSSCs depends mainly on the sensitizing ability of dyes, thus chlorophyll from *Elaeis Guineensis* which is more sustainable and non-toxic may replace the current synthetic dye photosensitizer, ruthenium (II) polypyridyl. The carbonyl group found via the FTIR analysis also elucidates that *E. guineensis* chlorphyll to have a strong anchor to the mesoporous semiconductor TiO_2 in DSSC where good adhesion is one of the best characteristics of an effective dye.

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APPENDIX

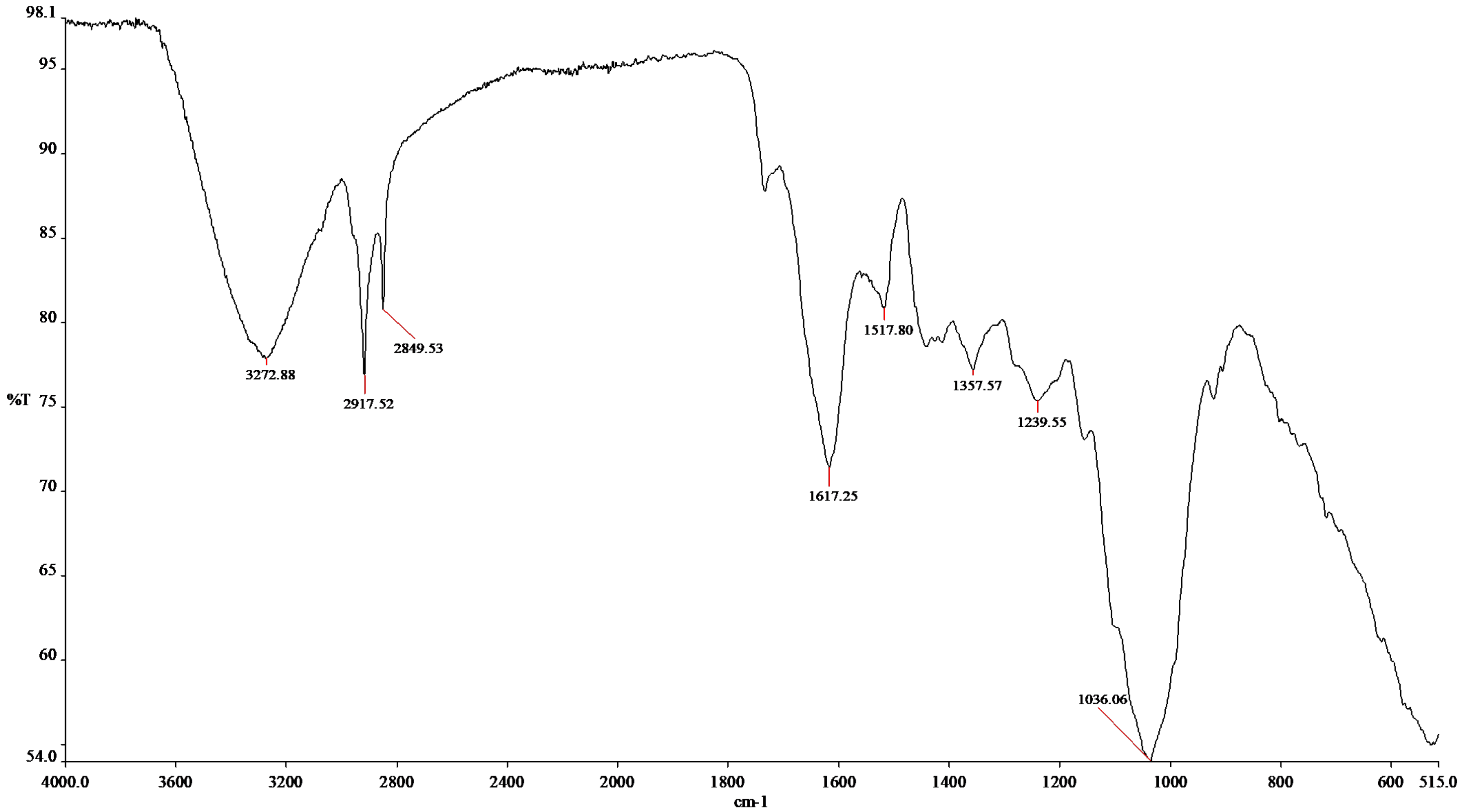

Fig 5: FTIR spectra of E. guineensis chlorophyll