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*by Harsasi Setyawati*

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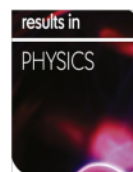
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## Effect of metal ion Fe(III) on the performance of chlorophyll as photosensitizers on dye sensitized solar cell



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### ABSTRACT

The energy crisis is a major problem facing the world today and will need a renewable energy source that is environmentally friendly; one of these is the dye sensitized solar cell (DSSC). DSSC is photochemical electric cell that can convert solar energy into electrical energy. This research aims to study the characteristics of chlorophyll compounds with the addition of metal ions Fe(III) and to determine the effect of Fe(III) on the performance of chlorophyll as a photosensitizer in the DSSC. The formation of complex compounds of Fe(III)-chlorophyll is shown by the phenomenon of metal ligand charge transfer (MLCT) at a wavelength of 263.00 nm and absorption transition d-d at 745.00 nm. Fourier transform infrared characterization of the binding of Fe-O complex compounds appears at  $486.06\text{ cm}^{-1}$ . The complex compound of Fe(III)-chlorophyll has a magnetic moment value of 9.62 Bohr Magneton (BM). The existence of ion Fe(III) in chlorophyll can improve the performance of chlorophyll as a dye sensitizer with a maximum current of  $4.00\text{ mA/cm}^2$ , maximum voltage of 0.18 volts and efficiency values of 1.35%.

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### Introduction

Energy is an essential requirement for human survival. Reliance on fossil fuels causes the supply of energy sources to dwindle while demand for energy increases [1]. The problems of the energy crisis have prompted many researchers to develop technologies that generate renewable energy. Solar energy is one type of renewable energy actively being developed at this time. Applications of dye sensitized solar cells (DSSC) using dye to capture sunlight have the potential to be developed because DSSC produces electricity and its applications can easily be applied to life [2].

DSSC technology is an attractive and economical source of renewable energy that converts solar energy into electrical energy. It was first discovered by Michael Grätzel and Brian O'Regan in 1991 [3]. DSSCs consist of semiconductors: photoanode, dye sensitizer, electrolyte and counter electrode [4]. DSSC performance is

very dependent on the material and the quality of the semiconductor electrode and the dye sensitizer used for the manufacture of the cells [5]. At the present time, the most efficient dye sensitizer used for photoinduction electron transfer in the DSSC is complex compound Ruthenium(II) polypyridyl, with an overall photovoltaic conversion efficiency of 10% [6]. However, because of the high costs and complicated synthesis process, researchers began developing dye sensitizers from transition metal that are cheaper to produce, yielding DSSCs that are more effective, environmentally friendly and low-cost [4].

DSSC utilizes the photoelectrochemical effects of dye in a number of materials, mainly organic materials. Dye used for sensitizer comes in synthetic and natural types. The use of natural dye as the sensitizer material has advantages including availability of abundant natural materials, low operating costs and a simple and easy to use extraction procedure that uses organic solvents without the need for high purity [7]. However, natural dye has the disadvantage of being easily degraded at high temperatures and less efficient when compared with synthetic dye. Previous studies have synthesized derivatives of 4,4'-dibenzodifuran-2,2'-bipyridine as

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a ligand for ruthenium dye, which, measured at AM 1.5G conditions, has a conversion efficiency of up to 21% [8]. In addition, it is known that if the produced current and voltage are high, the produced conversion efficiency is also higher.

A semiconductor material widely used in DSSC is  $\text{TiO}_2$  (titanium oxide), which has a mesoporous structure [9]. Titanium semiconductors have an energy gap of 3.2 eV and absorb light in the ultra-violet region. For application in DSSC,  $\text{TiO}_2$  is used commonly in the anatase phase because it has high photoactive ability. This material is chosen because it has advantages such as being inert, stable against photocorrosion, easily accessible, widely used and non-toxic [10].  $\text{TiO}_2$  is applied to the DSSC and should be prepared on broad surfaces so that more dye can be absorbed.  $\text{TiO}_2$  is expected to increase the photocurrent. In addition, the use of a dye capable of absorbing a wide spectrum of light and in accordance with the energy band of  $\text{TiO}_2$  [6]. One aspect that affects the performance of solar cell-based DSSC is the number of dye molecules that can be absorbed by the  $\text{TiO}_2$  layer. More dye molecules be changed to adsorbed then causes greater absorption of photons and produces a higher efficiency value. The number of dye molecules adsorbed on the  $\text{TiO}_2$  layer is influenced by the molarity of adsorbed dye solution when dipping the  $\text{TiO}_2$  layers in dye solution [11].

The dye or plant pigment used as photosensitizer in this study is chlorophyll powder of classic mulberry powder (CMP). Researchers have proven that chlorophyll and xanthophyll can be excited by the exposure of the dye. As a result of its development, researchers have gained a better energy conversion efficiency on derivatives of chlorophyll dye because it has a carboxylic group [12]. Chlorophyll as a major pigment is effective as a photosensitizer in the photosynthesis process of green plants, which have maximum absorption at 670 nm; thus, chlorophyll is a compound that is suitable for use as a photosensitizer [6]. However, the stability of chlorophyll is weak, and it easily decomposes when exposed to heat, light and acidic and alkaline conditions [13]. The performance of a photosensitizer is influenced by the level of absorption peaks; if the absorption peak of a dye is high, it indicates that more photon energy can be absorbed and converted into electrical energy in solar cell applications. The produced current and voltage also affect the performance of the photosensitizer: The higher I-V indicates that the produced efficiencies have also increased [14].

Chlorophyll as a photosensitizer produces a strong absorption peak at a wavelength of 420 nm and 660 nm. The chlorophyll extract shows open circuit voltage (Voc) of 0.585 mV, short circuit current (Isc) around 1.96 mA, a fill factor (FF) around 47% and conversion efficiency of 0.538% [15]. This shows that the conversion efficiency in natural dye is much lower when compared with synthetic dye. One technique to improve the photostability of chlorophyll can be done through interaction with metals to form complex compounds [16]. The inspiration for this technique came from the use of mordant by craftsmen of batik in Indonesia to increase the brightness and color durability of batik using natural dyes. Several types of mordants used include salt, chalk, alum ( $\text{Al}_2(\text{SO}_4)_3$ ) and  $\text{FeSO}_4$ . The coloring of batik using mordant yields brighter colors and greater resistance to water and heat [17]. The increase in brightness and color durability is expected due to the formation of complex compounds between the mordant and natural dyes. Therefore, in this study, we will assess the addition of salt  $\text{FeCl}_3$  on chlorophyll dye. In addition to increasing the stability of chlorophyll against light radiation, the addition of Fe(III) is expected to widen the light absorption area of natural dyes to the near-infrared spectrum region, which is the largest component of the sunlight spectrum.

Previous studies have addressed the formation of curcumin complex with Fe(III) and the antioxidant curcumin to improve photostability. Curcumin interacts with Fe(III) via the carbonyl group and provides better antioxidant photostability [16,18]. Interactions

that occur in complex molecules are characterized by the shift in wavelength bathochromic derived from electronic transition  $\pi \rightarrow \pi^*$  and  $n \rightarrow \pi^*$ . Chlorophyll also has a carbonyl group and is expected to have an interaction similar to curcumin when it binds to Fe(III).

This research studied the effect of ion Fe(III) on the performance of chlorophyll as a dye sensitizer in DSSC. Fe was selected because it is a transition metal in which the electron configuration d[6] is the same as ruthenium and osmium metals (used as dye solar cell), but ferrous metals are more easily obtained because of their abundance in nature when compared to other metals. It also has a quantum that is relatively high for producing sensitization on the nanocrystalline  $\text{TiO}_2$ ; it is cheaper and can be obtained in Indonesia more easily than other metals and has been studied previously as a complex for solar cells; it is soluble in polar solvents; and its wavelength in the UV-Vis is 551 nm [19].

The formed complex compound of Fe(III)-chlorophyll will be characterized using UV-Vis spectrophotometer, Fourier transform infrared (FTIR), magnetic susceptibility balance (MSB) and the electrical conductivity test. The absorption of sunlight in the DSSC cells made by dyes that are coated on the surface of  $\text{TiO}_2$  will be measured using the doctor blade technique, which is very simple and easy compared to other techniques. The doctor blade technique is a method of coating  $\text{TiO}_2$  suspension on semiconductor glass using a rod stirrer by rolling the stirrer on the glass surface to flatten the suspension of  $\text{TiO}_2$  that has been dropped [20]. The properties of the complex compound of Fe(III)-chlorophyll will be tested using measurements of produced current and voltage by multimeter when exposed to direct sunlight.

## 2.6 Materials and methods

### Materials and chemicals

The chemicals used in this study have a purity degree of pro analysis and include, among others: sodium hydroxide (NaOH), ethanol ( $\text{C}_2\text{H}_5\text{OH}$ ), hydrochloric acid (HCl), iron(III) chloride ( $\text{FeCl}_3$ ),  $\text{TiO}_2$  Degussa P25, acetonitrile, aquabidest, electrolite solution of  $\text{I}_2$  in KI and chlorophyll powder CMP (PT. Health Wealth International).

### Research procedure

#### Preparation of chlorophyll

A total of 1 g of chlorophyll powder was dissolved with aquabidest in a glass beaker, then transferred into a 10 mL volumetric flask up to the mark.

#### Saponification reaction of chlorophyll

A total of 2 g of NaOH were dissolved in 50 mL aquabidest. Five milliliters of NaOH solution then was diluted in 50 mL of aquabidest to obtain a concentration of 0.1 M NaOH solution. Two milliliters of this solution then were added to 8 mL ethanol to form ethanol-NaOH with total volume of 10 mL. As much as 5 mL of chlorophyll solution was added to the solution and homogenized for 2 h in a water bath at a temperature of 50 °C. The addition of NaOH formed a reaction with alkaline conditions. A total of 1 mL of chlorophyll was taken and dissolved in 50 mL aquabidest and then checked using the UV-Vis spectrophotometer 1800 Shimadzu at a wavelength of 639.00 nm [13].

#### The concentration optimization of complex compound Fe(III)-chlorophyll

The chlorophyll solution was combined with two concentrated drops of HCl to achieve a pH of 2–3. Furthermore,  $\text{FeCl}_3$  solution



with concentrations of 5%, 10% and 20% were added to get the optimum concentration conditions. The solution with each concentration was homogenized at a temperature of 60–70 °C for 1 h; then the solution was evaporated to one-third part and reheated until the filtrate was exhausted. Next, the precipitate was heated in an oven at a temperature of 80 °C for 24 h to form the product Fe(III)-chlorophyll [13].

#### The characterization of complex compounds of Fe(III)-chlorophyll

##### The characterization of complex compounds of Fe(III)-chlorophyll using UV-Vis spectrophotometer

One milliliter of chlorophyll solution was dissolved by aquabidest in a 50 mL volumetric flask. The absorbance was measured at wavelengths between 200 and 800 nm using the UV-Vis spectrophotometer [21]. The solution from saponification of 1 mL chlorophyll was dissolved in a 50 mL volumetric flask with aquabidest. The absorbance in the region 200–800 nm then was measured.

##### The characterization of complex compounds of Fe(III)-chlorophyll using FTIR

The characterization using FTIR aims to observe the formation of bonds between the metal with ligands and to identify the functional groups on the compound formed [4]. The complex compound of Fe(III)-chlorophyll was mixed with KBr; then the mixture was made into pellets, and IR absorption at wave number 4000–300  $\text{cm}^{-1}$  was measured [22].

##### The characterization of complex compounds of Fe(III)-chlorophyll using MSB

The characterization using MSB aims to determine the magnetic properties of complex compounds at room temperature. The empty MSB tube was weighed as  $m_0$ , and then the tube was inserted in the MSB tool to obtain the value of  $R_0$ . The MSB tube was filled with the formed compound and reweighed to obtain the value of  $m_1$ . The greatest value of sample was given a value of 1. The MSB tube of the sample was put back in the tool to obtain the value R [23].

##### The characterization of electrical conductivity of complex compound of Fe(III)-chlorophyll

This characterization aims to determine the properties of complex compounds of Fe(III)-chlorophyll that are dissolved in dimethyl sulfoxide (DMSO) at a concentration of  $10^{-4}$  M. The electrical conductivity of complex compounds will be compared with the electrical conductivity of DMSO using a conductometer.

#### Preparation of DSSC

##### Coating of $\text{TiO}_2$ on the glass plate

The glass plate was immersed in ethanol for 24 h to remove dirt. The glass plate then was dried. Compounds  $\text{TiO}_2$  Degussa P 25, as much as 0.1 g, were suspended in 100 mL of 98% methanol. The suspension of  $\text{TiO}_2$  was coated on the semiconductor glass measuring  $2.5 \times 2.5 \text{ cm}^2$ , and  $\text{TiO}_2$  then was deposited with the help of scotch tape on the side of the glass that has resistivity, thus forming an area of  $2 \times 2 \text{ cm}^2$ . Scotch tape serves to control the thickness of the suspension of  $\text{TiO}_2$ . The suspension of  $\text{TiO}_2$  was deposited over the place created by using the doctor blade method. The stirring rod or spatula was rolled on the surface of the glass to flatten the suspension of  $\text{TiO}_2$  dripped until it reached a certain thickness. Glass that had been coated with  $\text{TiO}_2$  was allowed to stand at room temperature for 45 min and then was heated at 450 °C for 30 min. It then was allowed to stand until the temperature reached 70 °C [11].

##### The characterization of $\text{TiO}_2$ bond and complex compounds of Fe(III)-chlorophyll using FTIR

The glass plate that had been coated with  $\text{TiO}_2$  was soaked in the complex compound of Fe(III)-chlorophyll for 24 h. The glass plate was filed with another glass plate to obtain  $\text{TiO}_2$  powder. The obtained powder was mixed with KBr to make pellets, and then IR absorption in the region of 4000–300  $\text{cm}^{-1}$  was measured.

##### Preparation of electrolyte solution

The electrolyte solution was made with a concentration ratio of 0.5 M iodide salt and 0.05 M  $\text{I}_2$ . A total of 0.498 g of KI was dissolved in 6 mL of acetonitrile in a glass beaker. As much as 0.076 g of  $\text{I}_2$  and 6 mL of acetonitrile were placed in another glass beaker, then stirred until homogeneous. The solution in both glass beakers was mixed and stirred [24].

##### Preparation of DSSC working electrode

The glass plate that had been coated with  $\text{TiO}_2$  suspension was immersed in complex compound of Fe(III)-chlorophyll on a petri dish for 24 h. After coating, the glass plate is stored in dark glass bottles to avoid sunlight and ensure optimal absorption. For the use in the long term, the glass is stored in sealed glass bottles to avoid scratches that can damage the surface of  $\text{TiO}_2$  [11].

##### Preparation of DSSC reference electrode

The glass plate was shaded evenly using a graphite pencil and then clamped and taped on the edges. It was then burned for 30 min using the flame from a candle to form a carbon layer [21]. The coating is intended as a catalyst to accelerate the reaction in the DSSC.

##### Preparation of DSSC tool

The working electrode that had been coated with  $\text{TiO}_2$  and soaked with complex compounds was placed on the table with a layer of complex compound of Fe(III)-chlorophyll in the top position. The reference electrode was placed opposite to the working electrode, and the electrolyte solution of  $\text{I}_2$  in KI was dripped between the two electrodes. The structure of the glass plate was clamped with binder clips on both ends of the part that was not coated to form a series of solar cells. The series of DSSC tools is shown in Fig. 1.

##### The measurements of current and voltage on the DSSC

DSSCs were connected by multimeter cables on both sides of the glass plate with the reference electrode at the positive pole and the working electrode at the negative pole. DSSCs that had been assembled were illuminated by direct sunlight with the

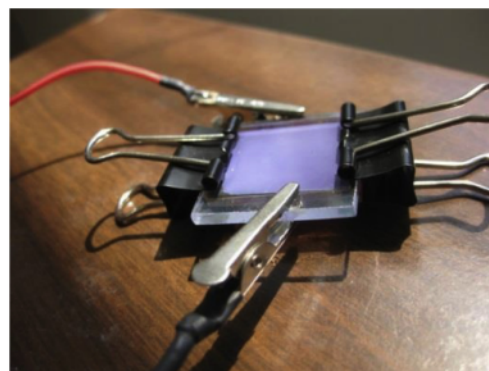


Fig. 1. The series of DSSC.

working electrode at the top; the maximum current and voltage then were measured. Output current was also measured by determining the active area of the solar cell [11]. The measurements of current and voltage are shown in Fig. 2.

#### The characterization of current and voltage on the DSSC using potentiometer

The characterization of current and voltage was done by connecting the DSSC and potentiometer that are adjusted in some way to determine the characteristics of current and voltage [21]. The measurement series of the I-V curve is shown in Fig. 3.

## 41 Results and discussion

In this study, we have investigated the characteristics of chlorophyll compounds that have been added to the metal ion Fe(III) and the effect of the presence of metal ion Fe(III) on the performance of chlorophyll, which is used as a photosensitizer in the DSSC. The complex compounds formed were characterized using UV-Vis spectrophotometer, FTIR, MSB and electrical conductivity test, which is measured with a conductometer. Furthermore, the performance of the photosensitizer was tested by measuring the current and voltage using a multimeter to better understand the effect of Fe(III) to the chlorophyll as ligand.

#### Saponification reaction of chlorophyll

The saponification reaction of chlorophyll, which aims to reduce the stability of chlorophyll to allow it to form bonds with other metals, was done by reacting chlorophyll that has been diluted with aquabidest with base. In the present study, the base used is ethanol-NaOH, which was homogenized in a water bath for 2 h [13]. The addition of bases aims to open the isocyclic ring on chlorophyll and remove the phytol group [25].

#### The determination of the optimum concentration of metal ion Fe(III) in complex compounds of Fe(III)-chlorophyll

The preparation of complex compounds of Fe(III)-chlorophyll was carried out by reacting Fe(III) derived from the compound  $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$  with chlorophyll solution that had been saponified with NaOH 0.1 M at a temperature of 50 °C. The various concentrations of metal ion Fe(III) used in this study were 5%, 10% and 20%.

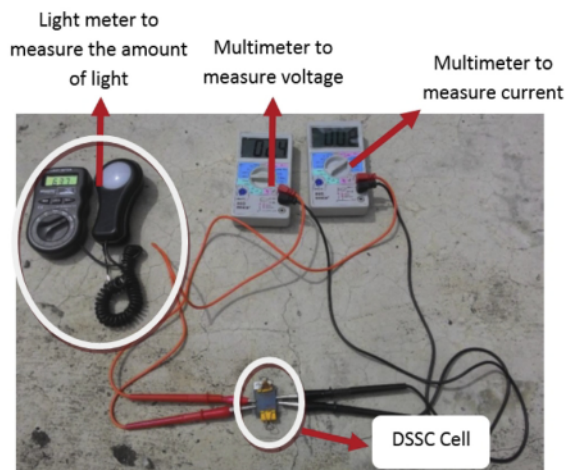


Fig. 2. The measurements series of current and voltage on the DSSC.

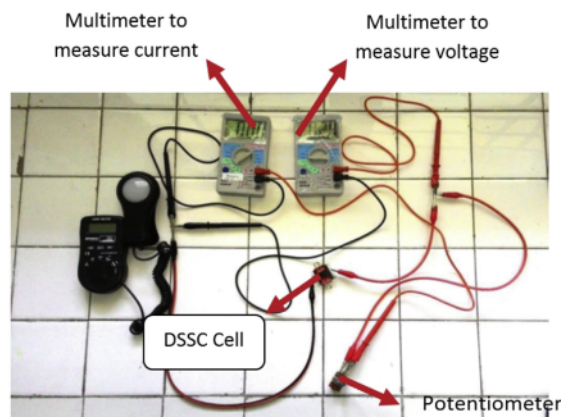


Fig. 3. The measurements series of I-V curve on the DSSC.

To 5 mL of saponified chlorophyll solution, we added 2 drops of HCl 37% to obtain pH 2–3. This is done to neutralize the saponified chlorophyll solution so that the metal ion Fe(III) is easily attached to the carbonyl group of the chlorophyll. Concentration optimization of the addition of metal ion Fe(III) was then conducted to obtain optimum results from the complex compounds formed. The 5 mL  $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$  solution with concentrations of 5%, 10% and 20%, respectively, were added to the saponification solution and heated at a temperature of 60–70 °C for 1 h. The filtrate was analyzed using UV-Vis spectrophotometer at a wavelength of 639 nm to analyze which concentration of metal ion Fe(III) is optimal.

Based on the absorbance data in Table 1, the optimum conditions for preparation of complex compounds is Fe(III) with a concentration of 10% because this concentration yielded the highest absorbance. In addition on the preparation of complex compounds can also be seen that Fe(III) with a concentration of 20% produces lower absorbance when compared with Fe(III) with a concentration of 10%. The reasons for this phenomenon may be a formation of complex with another composition or dimerization (aggregation) of reagent (ligand) or complex, high ionic strength can also influence on the activity of species or their electronic state, or will have dehydration and medium polarity effects. After obtaining the optimum concentration of Fe(III), filtrate of the solution was evaporated to one-third and then evaporated again until the entire filtrate turned into precipitate. The obtained precipitate then was put into an oven for 24 h at a temperature of 80 °C. After baking, the precipitate took the form of black powder. After the complex compound was successfully established, we then conducted characterization to determine the characteristics of the complex compounds. The solid of the complex compound of Fe(III)-chlorophyll is shown in Fig. 4.

#### The characterization of complex compound of Fe(III)-chlorophyll

The complex compounds that were made were characterized to determine their properties and characteristics. The characteriza-

**Table 1**  
The absorbance data for determination of optimum concentration on metal ion Fe(III).

Concentrations of metal ion Fe(III)	Absorbance
5%	0.006
10%	0.061
20%	0.049





Fig. 4. The solid of complex compounds of Fe(III)-chlorophyll.

tion was conducted using UV–Vis spectrophotometer, FTIR, MSB and conductometer.

#### The characterization of complex compounds of Fe(III)-chlorophyll using UV–Vis

The characterization using UV–Vis spectrophotometer was conducted to determine the maximum absorption wavelength in the wavelength range of 200–800 nm. The characterization results with UV–Vis spectrophotometer are shown in Table 2.

Table 2 shows that there is a difference between the maximum wavelength of the complex compound of Fe(III)-chlorophyll and chlorophyll. The maximum wavelength of the complex compound of Fe(III)-chlorophyll in UV and the visible spectrum is 263 nm and 745 nm, respectively, while the maximum wavelength of chlorophyll is 404 nm and 628.50 nm, respectively. The difference in maximum wavelength that emerged shows that the complex compound of Fe(III)-chlorophyll has been formed. The maximum wavelength that appears in the region of 200–300 nm is due to the phenomenon of charge transfer in the form of Metal to ligand charge transfer (MLCT). In the complex compounds of Fe(III)-chlorophyll, this phenomenon occurred because metal ion Fe(III) has lower oxidation states [26]. The wavelength of MLCT on complex compounds of Fe(III)-chlorophyll can be used as a dye sensitizer in DSSC due to the phenomenon of charge transfer. The maximum wavelength of the compound is also influenced by the transition d–d that appears in visible areas [27]. This transition changes the distribution of metal–ligand charge because the electron density shifts from orbitals with high metal character to high ligand orbitals. This phenomenon is included in the phenomenon of MLCT [28]. The MLCT phenomenon occurs because the ligand has orbital  $\pi^*$  with low energy, and the presence of MLCT phenomenon in the complex compound makes it very useful as a dye sensitizer because it can optimally absorb light for conversion into electrical energy. When a compound has many electrons in d orbitals and the ligand is abundant, it causes the compound to undergo the MLCT phenomenon [28].

Table 2

The characterization result of complex compounds of Fe(III)-chlorophyll and chlorophyll.

Compounds	Maximum wavelength (nm)		
Fe(III)-chlorophyll	263.00	–	745.00
chlorophyll	–	404.00	628.50

In this study, Fe(III)-chlorophyll was dissolved in dimethyl sulfoxide (DMSO) as a solvent because this solvent has a high degree of polarity. In addition, DMSO does not liberate hydrogen when met with very electropositive metal [29].

#### The characterization of complex compounds of Fe(III)-chlorophyll using FTIR

The characterization of complex compounds of Fe(III)-chlorophyll by FTIR determines the functional groups and the bond formed between metal and ligands. The differences in the FTIR spectra between chlorophyll as ligand and the complex compound of Fe(III)-chlorophyll indicates that the compound has been successfully synthesized. The characterization results of chlorophyll as ligand and complex compound Fe(III)-chlorophyll can be seen in Fig. 5; the FTIR data can be seen in Table 3.

Based on the FTIR spectra from Fig. 5 and FTIR data from Table 3, there is bonding between the metal Fe(III) and chlorophyll as ligand. This can be determined from the formation of Fe–O bond, so it can be concluded that the complex compound of Fe(III)-chlorophyll has been formed. The prediction bonding between the metal Fe(III) with chlorophyll as ligand is shown in Fig. 6.

#### The characterization of complex compound of Fe(III)-chlorophyll using MSB

The complex compound of Fe(III)-chlorophyll is characterized using MSB to determine its magnetism properties by measuring the magnetic moment of the complex compound. Based on Table 4, the complex compound of Fe(III)-chlorophyll has a magnetic moment of 9.62 Bohr Magnetron (BM). When viewed from the perspective of valence bonds, chlorophyll ligands are weak and cannot urge the electrons in the d orbitals of Fe(III) to pair [30]. In this study, chlorophyll used as ligand donated six lone pair electrons, which are unpaired in metal Fe(III) in orbitals s, p and d; the hybridization formed therefore is  $sp^3d^2$  with an octahedral shape. The magnetic properties of complex compounds of Fe(III)-chlorophyll–based valence bond theory is shown in Fig. 7.

Based on Table 4, complex compounds of Fe(III)-chlorophyll have a magnetic moment of 9.62 BM, which indicates that there are nine unpaired electrons, but Fig. 7 shows that there are only five unpaired electrons. This is because the chlorophyll as a ligand that is bound by metal also contributed to increase the magnetic properties, which also increases the magnetic value.

#### The electrical conductivity characterization of complex compound of Fe(III)-chlorophyll using conductometer

The characterization of the electrical conductivity of complex compounds via conductometer aims to determine whether the complex compound is molecular or ionic. Measurements of the electrical conductivity of the complex compounds are then compared with the electrical conductivity of DMSO, the solvent used. If the conductivity value of the complex compounds of Fe(III)-chlorophyll is higher than that of the solvent (DMSO), this indicates that the complex compound is an ionic compound. If the conductivity value of the solvent (DMSO) is higher than that of the complex compound, this indicates that the complex compound is

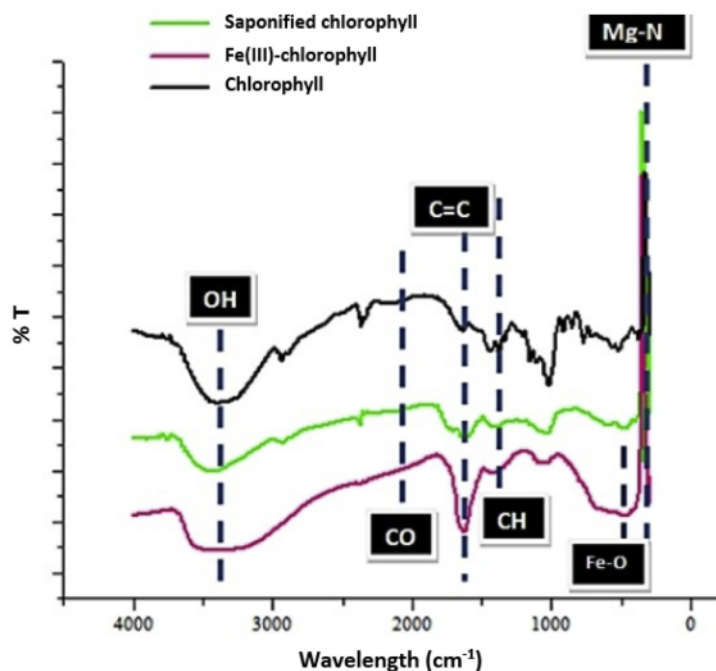


Fig. 5. FTIR spectra of chlorophyll, saponified chlorophyll and complex compounds of Fe(III)-chlorophyll.

Table 3

The FTIR data of ligand and complex compound.

Bond	Wavelength of ligand (cm <sup>-1</sup> )	Wavelength of complex compound (cm <sup>-1</sup> )	Wavelength Theoretically (cm <sup>-1</sup> )	References
Fe-O		486.06	430–480	Martak 2011 [9]
Mg-N	300.9	308.61	242–310	Mojumdar et al., 1998
OH	3394.72	3402	3362–3421	Ahmed et al., 2014
CO	2167.99	2167.99	2150–2200	Chakarova et al., 2011
CH	1381	1381	1350–1400	Bodirlau et al., 2007
C=C aromatic	1635	1620	1619–1640	Ahmed et al., 2014

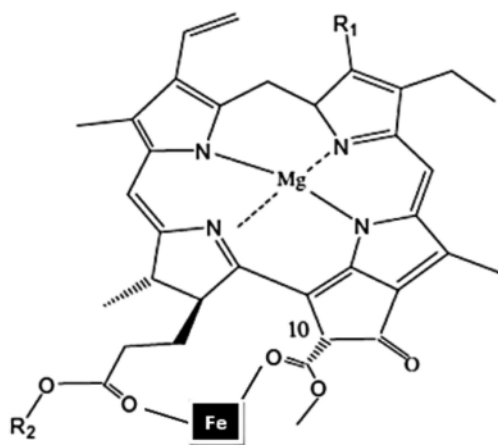


Fig. 6. The prediction bonding between the metal Fe(III) with chlorophyll as ligand.

a molecular compound. The conductivity values are shown in Table 5.

Based on Table 5, it can be seen that the conductivity of the complex compound of Fe(III)-chlorophyll is higher than that of

Table 4

The magnetic moment of FeCl<sub>3</sub>·6H<sub>2</sub>O, chlorophyll as ligand and Fe(III)-chlorophyll.

Compounds	Magnetic moment (BM)
FeCl <sub>3</sub> ·6H <sub>2</sub> O	5.13
Chlorophyll as ligand	0.81
Fe(III)-chlorophyll	9.62

the solvent (DMSO), indicating that the complex compounds are ionic. Ionic compounds easily conduct electricity because they have a high boiling point and melting point. Moreover, ionic compounds are easy to break down into ions because the electron transfer process can take place continuously. Ionic compounds are therefore good in DSSC applications [31].

#### Application of complex compounds of Fe(III)-chlorophyll in DSSC

In this study, before the complex compounds of Fe(III)-chlorophyll applied to DSSC, it needs to do some preparation. The preparation that has been done in this study such as: preparation of TiO<sub>2</sub> suspension, coating of TiO<sub>2</sub> on glass plate with doctor blade method, preparation of working electrode, preparation of reference electrode, and preparation of electrolyte solution. In the prepara-





functional groups on the ligand must have a lone pair of electrons that can bind with  $\text{TiO}_2$  [11].

Based on the spectra that can be seen in Fig. 11 at a wavelength of  $655.80 \text{ cm}^{-1}$ , Ti-O bonds are formed that show that complex compounds of Fe(III)-chlorophyll have bound to Ti. This finding agrees with the literature, which shows the bond between the Ti and O group of the ligand that appears at a wavelength of  $600\text{--}400 \text{ cm}^{-1}$  [32]. The prediction of chemical bonds of complex compounds of Fe(III)-chlorophyll with  $\text{TiO}_2$  is shown in Fig. 12.

#### The measurement of DSSC performance

The DSSC tool was assembled by placing working electrode and reference electrode in opposite directions on the glass plate. Drops of electrolyte solution were then placed between the two electrodes. The series was then clamped using clips, and efficiency measurements were taken using a multimeter. The series of DSSC are presented in Fig. 13.

#### The measurement of current

The measurement of current was done consistently for 14 days. The measurement was performed with variations of working electrode:  $\text{TiO}_2$ ,  $\text{TiO}_2$ -chlorophyll,  $\text{TiO}_2$  with complex compound of Fe(III)-chlorophyll and ion Fe(III). Measurement was performed on each DSSC to determine the current produced. In addition, we also compared the current produced in the complex compound with  $\text{TiO}_2$  and the mixture between Fe(III) and chlorophyll. This was done to determine the influence of the chemical bond between the complex compound and the ligand on the produced current. The measurement results are presented in Table 6 and Fig. 14.

Based on Fig. 14, it can be seen that the complex compounds of Fe(III)-chlorophyll have a higher current when compared to  $\text{TiO}_2$ , chlorophyll and mixture of metal Fe(III) with chlorophyll. This is because the complex compound of Fe(III)-chlorophyll exhibits the effect of metal that is chemically bonded, thus increasing the

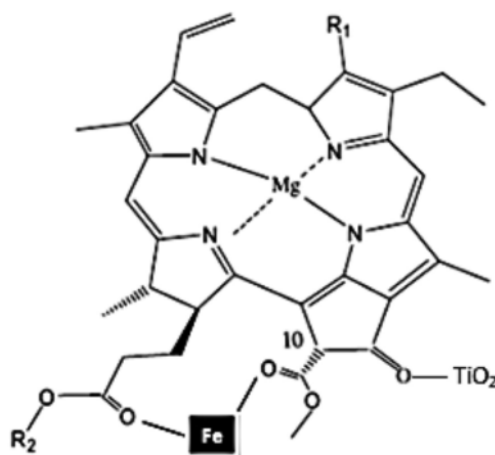


Fig. 12. The prediction of chemical bonds of complex compounds of Fe(III)-chlorophyll with  $\text{TiO}_2$ .

ability of the dye sensitizer [33]. In the complex compound, there is also conjugation bonds and the lone pair of electrons, which is easily excited and increases the produced current. The amount of current produced is also due to the intensity of sunlight, which is not fixed (i.e., it tends to fluctuate). If the intensity of sunlight is high, it increases the current produced; in contrast, if the intensity of the sunlight is low, it decreases the amount of current produced.

#### The measurement of voltage

The measurement of voltage aims to determine the voltage produced by DSSCs, which have been assembled with a variety of different compositions. The measurements were carried out for

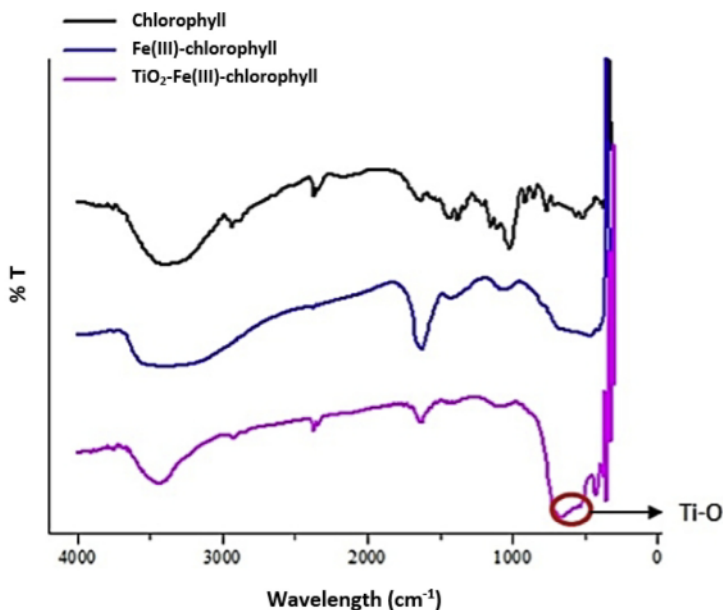


Fig. 11. The spectra of chlorophyll, complex compound of Fe(III)-chlorophyll and  $\text{TiO}_2$  with Fe(III)-chlorophyll.



Fig. 13. The series of DSSC.

14 days using various working electrodes: TiO<sub>2</sub>, TiO<sub>2</sub>-chlorophyll, TiO<sub>2</sub> with complex compound of Fe(III)-chlorophyll and TiO<sub>2</sub> with mixture of chlorophyll and ion Fe(III). The measurement results are presented in Table 7 and Fig. 15.

Fig. 15 shows that the complex compounds of Fe(III)-chlorophyll have voltage values that are relatively higher compared with TiO<sub>2</sub>, TiO<sub>2</sub>-chlorophyll and the mixture of Fe(III) and chlorophyll. The complex compounds of Fe(III)-chlorophyll have voltage value higher than those of the mixture of Fe(III) with chlorophyll, although the two compounds both contain metal ion Fe(III). The difference is that mixture of Fe(III) with chlorophyll

as ligand is not bound chemically, whereas the complex compound of Fe(III)-chlorophyll has interactions and chemical bonding that cause voltage value to increase. TiO<sub>2</sub>-chlorophyll also has higher voltage value when compared with TiO<sub>2</sub>, which indicates that the dye sensitizer in the solar cell can increase the voltage conversion capabilities over TiO<sub>2</sub>, which only serves as a semiconductor without dye.

40 The measurement of I-V curve

The determination of DSSC efficiency in converting sunlight into electrical energy is done by measuring the I-V curve. In this study, the measurement of electrical power in voltage produced by DSSCs was conducted by using potentiometer load with resistivity of 100 KΩ. The measurement results of current-voltage (I-V) are shown in Table 8 and Figs. 16–19.

This study carried out measurement of current and voltage of TiO<sub>2</sub>, chlorophyll as ligand, mixture of ion Fe(III) with chlorophyll and complex compounds of Fe(III)-chlorophyll and aims to determine the efficiency of each compound. From Figs. 16–19, it can be seen that the value of the short-circuit current (I<sub>sc</sub>) of TiO<sub>2</sub>, chlorophyll as ligand, mixture of ion Fe(III) with chlorophyll and complex compounds of Fe(III)-chlorophyll is 8.00 mA, 14.00 mA, 8.00 mA and 16.00 mA, respectively. We also obtained the value of open-circuit voltage (V<sub>oc</sub>) of TiO<sub>2</sub>, chlorophyll as ligand, mixture of ion Fe(III) with chlorophyll and complex compound of Fe(III)-chlorophyll: 0.10 V, 0.14 V, 0.12 V and 0.18 V, respectively. The maximum power point (mpp) is the point on the I-V curve that produces current-voltage and is obtained by determining the area

Table 6  
The current of working electrodes TiO<sub>2</sub>, TiO<sub>2</sub>-chlorophyll, TiO<sub>2</sub> with Fe(III)-chlorophyll and TiO<sub>2</sub> with mixture of chlorophyll and FeCl<sub>3</sub>·6H<sub>2</sub>O.

Days	Currents I <sub>sc</sub> (mA)				Lux
	TiO <sub>2</sub>	TiO <sub>2</sub> -chlorophyll	TiO <sub>2</sub> with Fe(III)-chlorophyll	TiO <sub>2</sub> with mixture of chlorophyll and FeCl <sub>3</sub> ·6H <sub>2</sub> O	
1	35.4	38.4	42.0	32.0	59.87
2	38.4	41.0	42.4	31.2	59.74
3	34.2	39.4	41.2	37.0	59.74
4	35.2	40.6	44.0	35.0	59.87
5	37.8	39.6	47.0	37.0	58.21
6	34.4	37.8	43.2	39.4	59.89
7	35.8	39.4	41.0	34.8	59.31
8	35.2	38.8	44.8	36.0	59.75
9	34.6	38.0	46.0	34.8	58.65
10	35.2	34.6	39.6	39.2	57.75
11	34.4	36.0	46.8	28.0	57.06
12	33.6	34.0	43.0	32.0	56.85
13	32.2	35.4	40.4	30.4	57.84
14	31.4	31.0	44.8	32.0	57.31

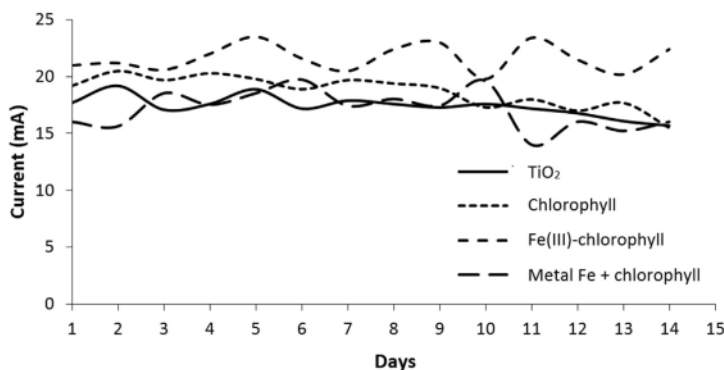
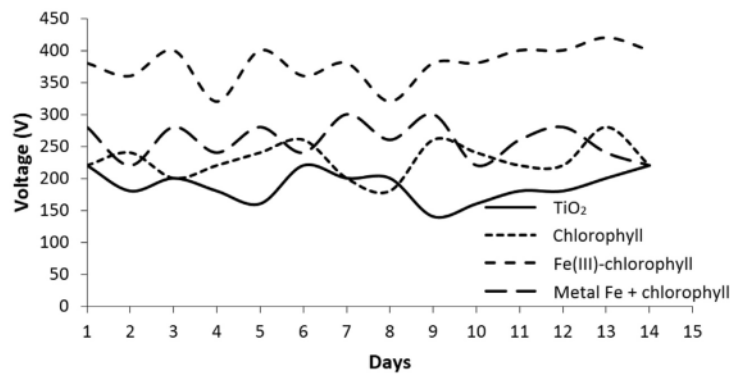


Fig. 14. The measurement result of current for 14 days.

**Table 7**The voltage of working electrode TiO<sub>2</sub>, TiO<sub>2</sub>-chlorophyll, TiO<sub>2</sub> with Fe(III)-chlorophyll and TiO<sub>2</sub> with mixture of chlorophyll and FeCl<sub>3</sub>·6H<sub>2</sub>O.

Days	Voltage (mV)				Lux
	TiO <sub>2</sub>	TiO <sub>2</sub> -chlorophyll	TiO <sub>2</sub> with Fe(III)-chlorophyll	TiO <sub>2</sub> with mixture of chlorophyll and FeCl <sub>3</sub> ·6H <sub>2</sub> O	
1	220	220	380	280	59.87
2	180	240	360	220	59.74
3	200	200	400	280	59.74
4	180	220	320	240	59.87
5	160	240	400	280	58.21
6	220	260	360	240	59.89
7	200	200	380	300	59.31
8	200	180	320	260	59.75
9	140	260	380	300	58.65
10	160	240	380	220	57.75
11	180	220	400	260	57.06
12	180	220	400	280	56.85
13	200	280	420	240	57.84
14	220	220	400	220	57.31

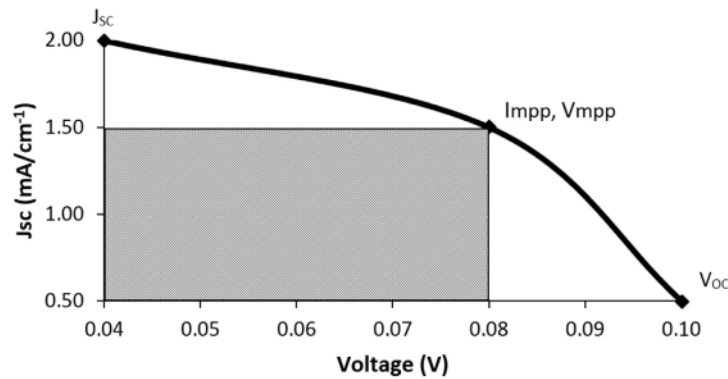
**Fig. 15.** The measurement result of voltage for 14 days.**Table 8**

The measurement results of current-voltage (IV).

Compounds	Voc (V)	Isc (mA)	Jsc	FF	$\eta$ (%)
TiO <sub>2</sub>	0.10	8	1.5	0.80	0.20
Chlorophyll	0.14	14	3.5	0.85	0.70
FeCl <sub>3</sub> ·6H <sub>2</sub> O + chlorophyll	0.12	8	2	0.83	0.33
Fe(III)-chlorophyll	0.18	16	4	1.12	1.35

under the curve. The mpp was obtained by determining the largest area yielded by multiplying the current by the voltage. The higher fill factor (FF) on the solar cell causes the produced efficiency value to also be higher [11].

From this study, it can be seen that the highest efficiency found was in the complex compound of Fe(III)-chlorophyll, equal to 1.35%. The efficiency results of complex compounds are higher when compared to chlorophyll as dye and the mixture of Fe(III)

**Fig. 16.** The I-V curve of TiO<sub>2</sub>.



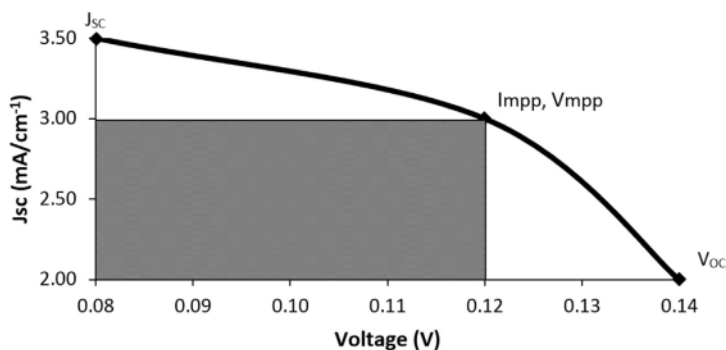


Fig. 17. The I-V curve of chlorophyll.

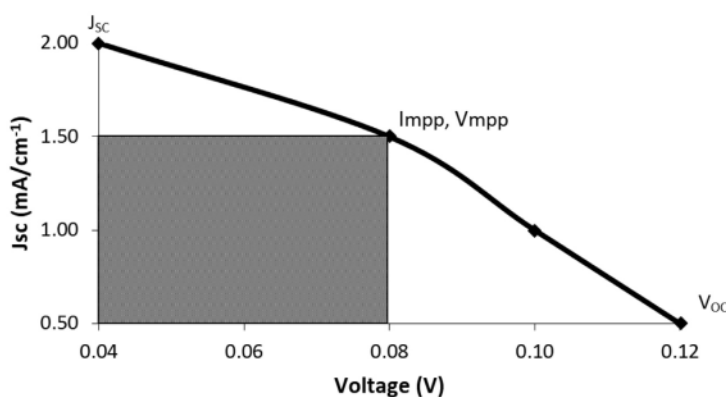


Fig. 18. The I-V curve of mixture of Fe(III) and chlorophyll as ligand.

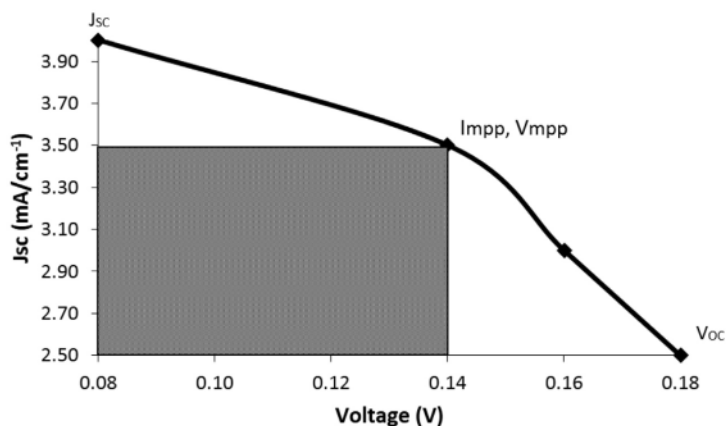


Fig. 19. The I-V curve of complex compounds of Fe(III)-chlorophyll.

with chlorophyll. This is because the MLCT phenomenon occurs in complex compounds of Fe(III)-chlorophyll, which causes more sunlight to be absorbed, which in turn increases the amount of sunlight that can be converted. The comparison of efficiency with previous research is presented in Table 9.

According to Table 9, the dye Fe(III)-chlorophyll solar cell has a higher efficiency value compared with chlorophyll from previous research. The complex compound of Fe(III)-chlorophyll has the advantage of being anionic compound that can undergo the MLCT phenomenon, has properties such as "collecting sunlight" and can

**Table 9**

The comparison of efficiency from previous studies.

Compound	Efficiency	References
Chlorophyll extract from spinach leaf	0.54%	Syafinar et al. (2015)
Chl- <i>e<sub>g</sub></i>	0.73%	Amao et al. (2014)
Iron formazan	4.22%	Listari and Akhlus (2010)
Chlorophyll	0.70%	The results of this current study
Fe(III)-chlorophyll	1.35%	
Mixture of FeCl <sub>3</sub> ·6H <sub>2</sub> O with chlorophyll	0.33%	

be used as photosensitizer in the DSSC. The electrons that are captured by the complex compounds cause excited electrons of complex compounds to move toward the conduction band of TiO<sub>2</sub>; the electrons then are transferred to the reference electrode and are received by the electrolyte solution. These electrons cycle through and in the presence of sunlight will be excited so that current that is produced can be used as a source of electrical power. With the advantage of the complex compounds that have been made in this study, we can see that adding metal ion Fe(III) improves the performance of chlorophyll in DSSC by improving efficiency as a photosensitizer in DSSCs. Moreover, the addition of metal ion Fe can cause chlorophyll performance to become more stable and not as easily degradable.

## Conclusion

The characterization of chlorophyll compounds with the addition of metal ion Fe(III) using UV–Vis spectrophotometer showed the presence of the MLCT phenomenon at a maximum wavelength of 263.00 nm. In the FTIR spectrum, the complex compounds showed the presence of Fe–O vibration of chlorophyll as ligand at a wavenumber of 436.06 cm<sup>-1</sup>. Analysis of magnetic properties using MSB obtained a magnetic moment value of 9.62 BM. This shows that the complex compound of Fe(III)-chlorophyll is paramagnetic. Based on the electrical conductivity test, it can be seen that the complex compound is an ionic compound. Moreover, the presence of Fe(III) on chlorophyll can increase the efficiency of chlorophyll as a photosensitizer in the DSSC. This can be seen from the increase in the voltage and current conversion value so that the produced efficiency becomes higher at 1.35%.

## Competing interests

The authors declare no competing financial interests.

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