

Original Article

Potential Application of Chlorophyll from *Syzygium paniculatum* as Electrode Dye in Solar Cells

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Abstract - This study aims to determine the chlorophyll content and chlorophyll dye on the nano TiO₂ electrode morphology. Color variations and good chlorophyll storage are dyes for coloring Nano TiO₂ electrodes on solar cells. The ingredient for making chlorophyll is the leaves of the *Syzygium paniculatum* plant. To prove that red shoot leaves' chlorophyll content is high, we will compare it with papaya leaves (*Carica papaya*) and cassava leaves (*Manihot utilissima*). This experiment uses a variation group with three experimental variables. The first variable is the variation of *Syzygium paniculatum* leaves with red color, green color, and a 50% red and 50% green mixture, as a comparison using cassava leaves and papaya leaves. The second variable is the solvent composition consisting of distilled water: chloroform: and 96% ethanol. The first solvent composition, in sequence, namely 3: 2: 1, and the second solvent composition, namely 3: 2: 1.5. The third variable is the storage time, 24 hours, 18 hours, and 12 hours. The results observed were chlorophyll levels, chlorophyll b levels, total chlorophyll levels, and the Nano TiO₂ electrode with chlorophyll dye. The results showed that leaf color and solvent composition significantly affect chlorophyll content and the electrode coating morphology. The results showed that the green leaf color and the 3: 2: 1,5 solvent compositions had the highest chlorophyll content, namely chlorophyll-a of 32.548 mg/L, chlorophyll b of 56.327 mg/L, and total chlorophyll of 88.750 mg/L. Moreover, the morphology with EDX analysis showed C, O, Ti, and Mg at 12 hours of storage. This study recommends using green leaf color from *Syzygium paniculatum* leaves with a solvent composition of 3:2:1.5 for DSSC.

Keywords - Cassava leaves, Chlorophyll dyes, Nano TiO₂, Papaya leaves, *Syzygium paniculatum* leave.

1. Introduction

As a natural pigment composed of chlorophyll-a and chlorophyll-b, chlorophyll plays an essential role in plants' green appearance. The chlorophyll structure combines four pyrrole rings to form a scaffold with a central magnesium ion and a long phytol chain. Chlorophyll-a consists of a methyl group (CH₃) in the side chain and tends to absorb more red light from the visible spectrum. Its chemical formula is known to be C₅₅H₇₂O₅N₄Mg with a molecular weight of 893 and turquoise. Chlorophyll-b consists of an aldehyde group (CHO) in the side chain and absorbs more purple-blue light from the visible spectrum. Its chemical formula is C₅₅H₇₀O₆N₄Mg, with a molecular weight of 907. Although chlorophyll-a is less polar than chlorophyll-b, they are insoluble in water and very soluble in ethanol and methanol. In higher plants, chlorophyll generally comprises 0.6 to 1.2%-wt leaf weight in dry matter. Chlorophyll absorbs the red and blue-purple regions of the solar spectrum. However, the green light does not drink but reflects, giving chlorophyll its green colour [1-4].

In photosynthesis in plants, chlorophyll molecules get light, which converts light energy into chemical energy.

Carotenoids are pigments that belong to the category of tetraterpenoids, which are components of photosynthesis. Plants absorb blue-green light and increase the range of sunlight used for photosynthesis. The demand for chlorophyll increases with growing awareness of using chlorophyll as a dye used as a natural sensitizer (as a light-capturing unit). Although it has potential as a sensitizer due to its unique chlorophyll content, the *Syzygium paniculatum* plant is only grown as a popular ornamental plant. In many public places in Indonesia, you can find this plant. Therefore, it would be wonderful to research this plant because of its chlorophyll content potential and produce it to meet market demand.

In this case, two factors are required in an economically viable process to make chlorophyll production. The first factor is the material with high pigment content, and the second is the efficient extraction mechanism [5-10]. This experiment will extract chlorophyll from the *Syzygium paniculatum* plant because no research has been conducted on this leaf extraction as a coloring agent. This study will validate the differences in chlorophyll extraction methods and chlorophyll purification techniques. Furthermore, a



comparison using chlorophyll from cassava leaves (*Manihot esculenta*) and papaya leaves (*Carica papaya*). Using cassava leaves and papaya leaves because they have an intense green colour indicates a high pigment content.

It tests chlorophyll levels using a UV Vis spectrophotometer and morphological observations using SEM (Scanning Electron Microscopy) to determine whether the electrodes have absorbed chlorophyll. Chlorophyll contains, among others, magnesium and calcium. Calcium is the most abundant intracellular ion in plants, essential for ion balance, high efficiency of photosynthesis, enzymatic activity, stress response, and water movement. Maintaining high potassium status is very important for plants under conditions of high levels of stress, such as salinity, drought, etc. [11-14]. This test determines the active layer's morphology, especially the donor and acceptor properties (e.g., solubility, crystallinity, and solubility), film processing, device configuration, and so on [22].

1.1. Botani of *Syzygium Paniculatum*

The redbud plant is a leafy shrub that is always green throughout the season. This plant has several local names, namely Pokok Kelat Paya (Malaysia), Chinese Red-Wood (Chinese), Wild Cinnamon, Red-lip, Australian Brush Cherry, and dan Kelat Oil. This redbud plant consists of roots, stems, leaves, flowers, seeds, or fruit. The shape of the red leaves is oval with a sharp point at the tip of the plate, for the structure of the redbud leaves has a leaf bone growing on each branch. The red leaves' colour is unique because the leaves will be red if the leaves are young. Over time the leaves will turn green so that the two colours combined make this plant looks beautiful.

The redbud plant has round, slightly flattened fruit about 0.7 cm in diameter, which in the middle of the upper surface of the fruit has a hollow, sweet taste with a distinctive aroma, and the fruit becomes shiny black when it is old. Figure 2 presents the fruit of the redbud plant [16].

1.2. Natural Dye

The process of photosynthesis in plants shows the presence of compounds in plants for dye. These substances are in the leaves or fruit, namely anthocyanins, xanthophylls, and chlorophylls. Researchers have proven that chlorophyll and xanthophyll can be excited by irradiating the dye [17]. (Zhang et al., 2018). The redbuds of the plant get their color from their anthocyanin content. The formation of anthocyanin and chlorophyll coincides. However, the anthocyanin content in the bud part happened to be more dominant than the chlorophyll content. Even though this anthocyanin's content is not greater than the chlorophyll content in redbud leaves as a whole [16].

Photo-sensitizing dyes play a critical role in DSSC performance in light absorption, exciton generation, and

electron injection into electron acceptors, determining short circuit current density and, thus, efficiency in DSSCs. Natural dyes are relatively low-cost materials due to the simple, straightforward processing to extract the stains from various vegetation sources. It is important to note that the natural dyes' pigments are the sources to determine the active photo-absorption spectral window [6].

2. Materials and Methods

2.1. Preparation of Chlorophyll Extract Solution

This part consists of materials preparation and the testing method used in the experiment. Make a dye solution with three colour variations of red shoots, namely dark green, red, and a green-red mixture. This study compared the red shoot leaves with papaya and cassava leaves.

The first step in making chlorophyll solution is to mix distilled water. It leaves with a plate composition of 50 grams: 50 ml of distilled water into a blender to form a perfectly homogeneous solution. Prepare six samples from each variation of the solution. Then reflux the acceptable leaf solution into a three-neck flask with a stirrer.

Next, add distilled water, ethanol, and chloroform to the flask with two variations of the solvent composition: 3:2:1 and 3:2:1.5. At this stage; it is heated for 10 minutes at a temperature of 60°C. Macerate the product by pouring it into a beaker. Furthermore, aluminium foil is coated on the outside of the beaker glass until the entire surface is covered. Doing so is to prevent the evaporation of ethanol and protect the solution from light damage.

Pour the extract into a beaker and store it in a dark place with three storage variations, namely 24 hours (No. 1), 18 hours (No. 2), and 12 hours (No. 3). This leaf maceration forms two layers, namely dark green on the bottom and brownish-green at the top. The extract was then filtered to separate the dregs' solution using an Erlenmeyer buncher funnel and a suction machine. Enter the resulting solution into a separating funnel and let it stand for a while to separate the chlorophyll from the solvent solution. The final result of the chlorophyll extract solution is ready to be used to soak the ITO electrode coated with a mixture of TiO₂ - PEG solution with a ratio of 1:4.

2.2. Testing Phase

Test the results of the redbud leaf extract solution using a UV-VIS spectrometer. This test determines the extract's sensitivity to the absorbed wavelength. Before carrying out the test, note calibrates the spectrometer machine with the solvent solution as a reference. Inserting the extract solution into the cuvette up to three-quarters of the cuvette, inserting it into the spectrometer, and setting up the spectrometer machine to determine the absorbance value 663nm and 645nm wavelengths.

3. Results and Discussion

3.1. Mathematical Equations

Record the results and calculate the chlorophyll content, using the following equation to determine the pigment using 96% ethanol according to reference [23].

$$\text{Total Chlorophyll} = (20.31 \times A_{645\text{nm}}) + (8.05 \times A_{663\text{nm}}) \quad (1)$$

$$\text{Chlorophyll} = (12.72 \times A_{663\text{nm}}) - (2.59 \times A_{645\text{nm}}) \quad (2)$$

$$\text{Chlorophyll} = (22.94 \times A_{645\text{nm}}) - (4.67 \times A_{663\text{nm}}) \quad (3)$$

This part will elaborate on dye absorbance characteristics using the UV - VIS spectrometer and the coating surface morphology analysis resulting from SEM.

3.2. Chlorophyll Extract Solution (Dye) Characteristic

Test the dye's chlorophyll content at a maximum wavelength of 663 nm and 645 nm using the UV - VIS -BEL Photonics LGS 53 Spectrophotometer. The analysis of variations in the solvent composition showed that ethanol solvent concentration treatment affected chlorophyll levels. Table 1 shows the test results (3: 2: 1 solvent composition) and Table 2 (3: 2: 1.5) solvent composition. Table 1 shows that the highest value of chlorophyll-a content is the leaves of SP-Green chlorophyll of 30,499mg/L, chlorophyll-b 55,144 mg / L, and total chlorophyll of 85,522 mg/L. While the lowest content is in SP-Red leaves for chlorophyll 9,629 mg/ L, 15,793 mg/L chlorophyll b, and 25,387 mg/ L total chlorophyll.

The data in Table 2 also shows the results that are not much different. The table shows that the highest chlorophyll content is SP-Green leaves, chlorophyll a 32,548 mg/L, chlorophyll b 56,327 mg/L and total chlorophyll 88,750 mg/L. In contrast, the lowest content was in the leaves of SP-Red with chlorophyll a 9.367 mg/L, chlorophyll b 14.878 mg/L, and chlorophyll-total 24.212 mg/L.

Table 1 and Table 2 show that the red leaves (*Syzygium paniculatum*) with red leaves are smaller than those in green. This condition is because the red leaves receive more sunlight, but less light is absorbed than the dark green leaves because the chlorophyll pigment content in the red leaves is less, so the light received is not fully absorbed and reflected. The lowest rebuds chlorophyll content was still higher than the papaya and cassava leaves.

From the results above, the higher the ethanol solvent the concentration used, the greater the chlorophyll content produced. These results are in line with research on the

effect of ethanol concentration on chlorophyll content, which states that the higher the use of ethanol solvent for extraction, the greater the cells' destructive power. The more compounds extracted, and the yield is higher. The chlorophyll-a content is lower than chlorophyll-b because chlorophyll-a is unstable to heat.

It changes faster to pheophytin a by 5-6 times compared to the rate of changing chlorophyll b to pheophytin b. The release of magnesium from chlorophyll is nine times more immediate than chlorophyll b. This condition's cause is the chlorophyll structure, which does not contain the formaldehyde (-CHO) group as in chlorophyll b. The higher the ethanol solvent concentration used in the maceration process, the higher the measured chlorophyll b content.

However, the high solvent concentration causes the extracted chlorophyll to be large. Still, if the solvent's interaction occurs at high temperatures, it will damage the chlorophyll compound. At high temperatures, the chlorophyll content in the extract is lower. The reason is that there are more unwanted organic compounds (impurities) in the extraction process, while the presence of thermal degradation causes chlorophyll to be damaged. The degradation of chlorophyll is called the pheofitination reaction. This reaction will result in the loss of magnesium in chlorophyll so that the colour changes to brownish green.

The results of the chlorophyll stability test on storage time in Table 1 and Table 2 with storage times of 24 hours (no.1), 18 hours (no.2), and 12 hours (no.3) indicate that the faster the storage time, the more chlorophyll produced. Or in other words, increasing the storage time will reduce the chlorophyll concentration in the extract. This condition occurs because chlorophyll is a straightforward compound (degraded) into its derivatives after processing.

Chlorophyll degradation continues until the product becomes colorless and begins with a gradual change from green to yellowish. The mechanism of the chlorophyll degradation reaction in plants causes the magnesium enzyme to catalyze the ester bond between the propionic acid residue in the macrocyclic ring and the phytol, causing the loss of Mg^{2+} ions.

In general, the chlorophyll degradation reaction takes place in two ways. The first reaction pathway is the change of chlorophyll-a to chlorophyllide-a in the presence of the chlorophyllase enzyme. Meanwhile, the second reaction pathway occurs due to the enzyme magnesium *dechelatase*, which converts chlorophyll-a into pheophytin-a. These two reaction pathways will produce pheophorbide-a, formed due to chlorophyllase pheophytin-a or Magnesium *dechelatase* from chlorophyll-a.

Table 1. Dye absorbance and chlorophyll level with solvent composition of 3:2:1

Sample	Absorbance		Chlorophyll a (mg/L)	Chlorophyll b (mg/L)	Total chlorophyll (mg/L)
	663 nm	645 nm			
SP ^a -Red	0.906	0.833	9.367	14.878	24.212
	0.987	0.881	10.273	15.601	25.838
	1.238	1.063	12.994	18.604	31.555
SP- Green	1.900	1.650	19.895	28.978	48.807
	2.558	2.088	27.130	35.953	62.999
	3.012	3.017	30.499	55.144	85.522
SP-Mix	1.220	1.225	12.346	22.404	34.701
	1.791	1.513	18.863	26.344	45.147
	2.032	1.955	20.784	35.358	56.064
CP	0.750	0.745	7.610	13.588	21.168
MU	0.754	0.702	7.773	12.583	20.327

Note: ^a) SP stands for *Syzygium paniculatum*; ^b) CP stands for *Carica papaya*; ^c) MU stands for *Manihot esculenta*

Table 2. Dye absorbance and chlorophyll level with solvent composition of 3:2:1.5

Sample	Absorbance		Chlorophyll a (mg/L)	Chlorophyll b (mg/L)	Total chlorophyll (mg/L)
	663 nm	645 nm			
SP-Red	0.936	0.879	9.629	15.793	25.387
	1.050	0.980	10.818	17.578	28.356
	1.406	2.003	12.697	39.383	51.999
SP- Green	1.813	1.935	18.050	35.922	53.895
	2.900	2.870	29.455	52.295	81.635
	3.191	3.105	32.548	56.327	88.750
SP-Mix	2.110	1.641	22.589	27.791	50.314
	2.350	1.870	25.049	31.923	56.897
	2.513	1.967	26.871	33.387	60.179
CP	1.231	1.092	12.830	19.302	32.088
MU	1.205	1.065	12.569	18.804	31.330

Note: ^a) SP stands for *Syzygium paniculatum*; ^b) CP stands for *Carica papaya*; ^c) MU stands for *Manihot esculenta*

Furthermore, pheophorbide-a will undergo a dioxygenase reaction to become a fluorescent compound and colourless Rusty Pigment 14 (Heaton & Marangoni 1996; Karolina et al. 2020). The chlorophyll content results in a good variety of several of these sample tests, followed by soaking TiO₂ as a dye. Then perform SEM testing on electrodes coated with the TiO₂-PEG ratio of 1:4.

3.3. Scanning Electron Microscope (SEM)

In general, to identify the types of atoms on a surface that contain multiple traces, most researchers use the EDS (Energy Dispersive Spectroscopy) technique. Most SEM tools have this capability, but not all SEMs have this feature. EDX is the result of X-rays, that is, by firing the X-ray at the position where you want to know the composition. After being fired at the desired position, specific peaks

representing the elements contained will appear. With EDS, we can also create elemental mapping by giving different colours to each piece on the material's surface and observing the morphology of dye extracted (SP-Green) coating on Nano TiO₂ using a Scanning Electron Microscope (SEM). Based on SEM testing, the results are shown in the following Figure 1 and Figure 2.

Figure 2 shows the dominance by elemental Oxygen (O) is the result of EDX in the line analysis process at a magnification of 20.00 K X; in the 9 μm region, it reaches a peak of approximately 200 cps. Most of the element titanium is still bound to oxygen, and only a small proportion is attached to carbon, namely in the 5 μm region at 24 cps.

EDX verifies the phases rich in Ti, C, and O in the microscope. This phase consists of small, agglomerated

plates with a diameter of 10 nm but cannot highlight the distances between layers. This condition may be due to the layer immediately disintegrating under the electron beam [21].

Figures 1 and Figure 2 show the results of SEM characterization of the TiO₂ 1000 nano (1 μm) nanoelectrode using SP-Green chlorophyll dye, which both contain Ti (Titanium), C (Carbon), and O (Oxygen) elements. Figure 1 shows the Mg elements' presence, while Figure 2 does not because the storage is too long to damage the Mg content. This chlorophyll decomposition process can occur either when chlorophyll is still in the plant cell (due to the chlorophyllase enzyme) or when it has been dissolved (after the addition of chloroform). This condition occurs due to the release of the bond compound at the Mg atom at the center.

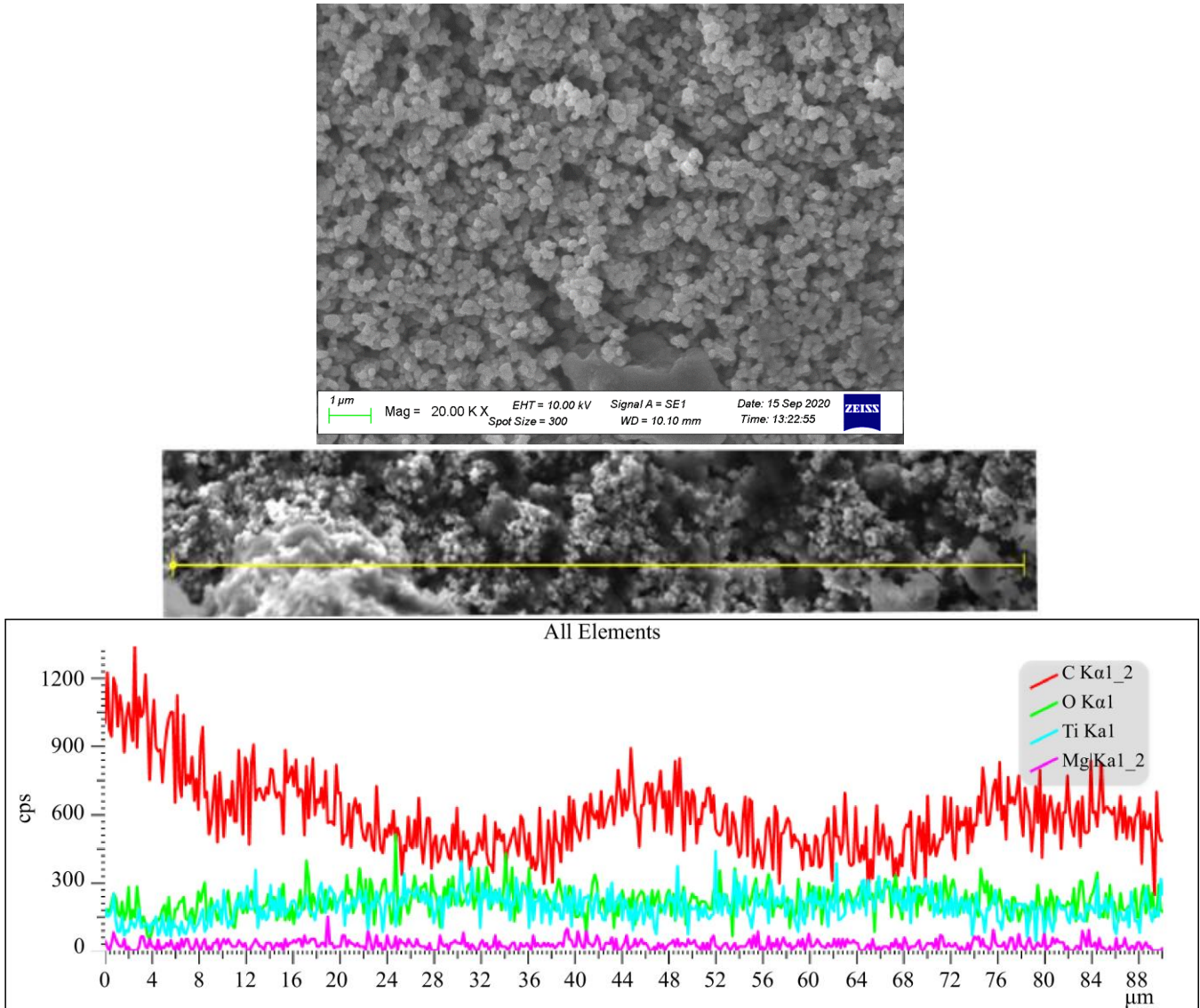


Fig. 1 EDX analysis results for nano TiO₂ electrodes with SP-Green chlorophyll dye of 6 hours storage

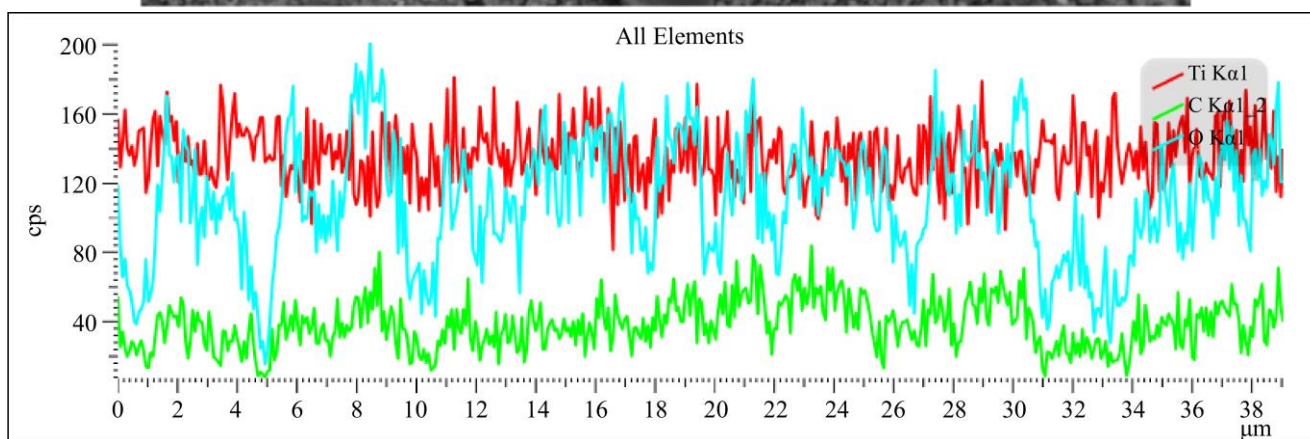
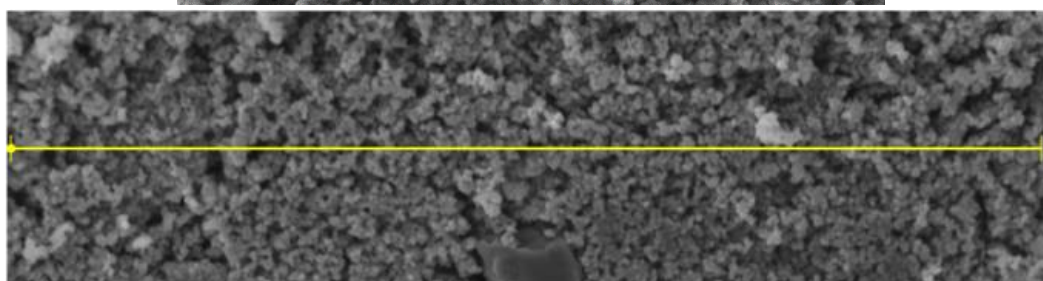
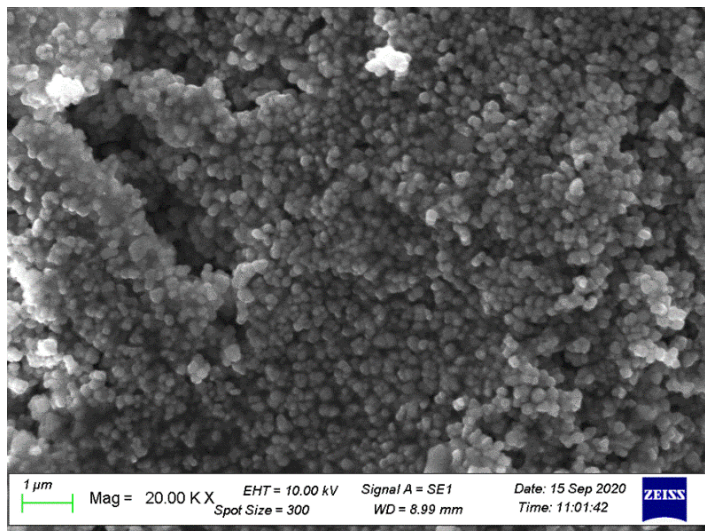


Fig. 2 EDX analysis results for nano TiO₂ electrodes with SP-Green chlorophyll dye of 24 hours storage

4. Conclusion

The conclusions based on the results of the chlorophyll absorbance test and the SEM-EDX test are as follows:

- Variation of the solvent ratio (aqua dest: chloroform: ethanol 96%) produced the best chlorophyll content with a 3:2:1.5 ratio.
- From the comparison of chlorophyll content, the highest yield is green leaves (SP-Green).
- The maximum chlorophyll content found were chlorophyll a 32,548 mg/L, chlorophyll b 56,327 mg/L and chlorophyll-total 88,750 mg/L. The lowest chlorophyll content was chlorophyll-a at 9,367 mg/L,

chlorophyll-b at 14.878 mg/L and chlorophyll-total at 24.21 mg/L.

- The best variation of storage time is 12 hours, where the storage time of 12 hours results in the remaining Mg elements, while none of the Mg elements remains at 24 hours of storage.
- Dominance by elemental Carbon (C) at storage time of the extract for 12 hours from line analysis at a magnification of 20.00 K X. The highest value was 1300 cps. At the time of the study, the EDX line was in an area of 3.2 μm. While the storage time for 24 hours only reached 200 cps in the 9 μm area.

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Contributions

SW conceived and designed the research — SW, AKS, and KH experiment. SW and AKS analyze the data. SW and KH wrote the manuscript. All authors have read and approved the manuscript.

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