ISSN (Print) : 2337-9030 ISSN (Online) : 2685-2608
PILLAR OF PHYSICS
JURNAL BERKALA ILMIAH FISIKA
Vol. 17 (1), 2024, page. 11-19
DOI : 10.24036/14238171074

EFFECT OF ADDITION OF SEAWEED (Sargassum sp) CHARCOAL WITH PURE GRAPHITE ON THE OPTICAL PROPERTIES OF GRAPHENE OXIDE SYNTHESIZED BY THE MODIFIED HUMMER'S METHOD

Rega Kurniawati [1], Ramli [1*], Gusnedi[1], Rahmat Hidayat[1]

[1] *Department of Physics, Universitas Negeri Padang, Jl. Prof. Dr. Hamka Air Tawar Padang 25131, Indonesia*
Corresponding author. Email: ramli@fmipa.unp.ac.id

ABSTRACT

Synthesis of graphene oxide from a mixture of pure Graphite and seaweed charcoal using a modified Hummers method was carried out with five variations of the composition, namely 100% graphite, 70% graphite–30% seaweed, 60% graphite – 40% seaweed, 50% - 50%, 100% seaweed. From this experiment, it will be seen how adding seaweed with Graphite affects the optical properties of the resulting graphene oxide. Characterization was carried out using FTIR, XRD, and SEM, and for optical properties, a UV-Vis Spectrophotometer was used. The FTIR test results showed the presence of carbon (C), hydrogen (H), and oxygen (O) functional groups. The XRD test results showed the crystal size of graphene oxide, and the SEM test showed graphene oxide's morphology in the form of thin sheets and chunks. The FTIR, XRD, and SEM tests showed that adding seaweed with Graphite had no effect. The results of the UV-Vis Spectrophotometer test showed that the highest absorbance value was at a variation of 50% - 50%, namely 49.547 at a wavelength of 245 nm, while for the lowest energy gap value, namely the variation of 100% seaweed 2.2875 eV and the highest 100% graphite 4, 2393 eV, the energy gap shows that there is an influence, the more seaweed composition used, the lower the energy gap.

Keywords : Graphene Oxide, Optical Properties, UV-Vis Spectrophotometer, Modified Hummer Method

I. INTRODUCTION

Indonesia is a maritime country with 2/3 of its territory consisting of oceans and has the longest coastline, ± 80,791.42 Km [1]. Around 27.2% of Indonesian waters are filled with flora and fauna species worldwide, and seaweed is one of the marine biotas with the most significant volume in Indonesian waters, around 8.6% of the total marine biota [2]. Sargassum sp contains bioactive ingredients: alginate, fucoidan, fucoxanthin, and phlorotannin [3]. Besides that, it also contains active compounds of steroids, alkaloids, phenols, and triterpenoids, which function as anti-bacterial, antiviral, and anti-fungal [4]; phenolic components [5], phlorotannins are the most common type of phenolic component found in brown seaweed, ranging from 0.74% to 5.06% [6]. The carbohydrate content of seaweed waste is 13-15% and has 16-20% cellulose components and 7-8% lignin [7]. Graphene oxide is made from a mixture of Graphite and seaweed-activated carbon. Carbon activation uses KOH compounds; previously, carbon was obtained by drying and burning brown seaweed samples at 300℃. Use a temperature of 300℃ because, at this temperature, the best carbon is obtained from burning seaweed. Graphene has high electrical and thermal conductivity; because of this, many scientists argue that graphene can store energy for applications such as batteries, solar cells, and supercapacitor cells [8].

Graphene is a carbon allotrope with a two-dimensional shape and hexagonal bonds [9]. According to [10], graphene oxide (GO) is oxidized graphene with bonds with a cluster function. There are two main problems in obtaining graphene; first, how can we produce graphene sheets on a sufficient scale? Even though graphite is cheap and available in large quantities, graphite is not easily peeled off to produce single-layer graphene sheets. Moreover, the second problem is that graphene sheets are challenging to combine and distribute homogeneously into various application matrices. The solution to this problem is that GO, which contains many oxygen-based groups, can be obtained easily from graphite oxidation. GO is a compound

derived from graphene, which contains carbon, oxygen, and hydrogen. So, researchers are interested in making graphene oxide from pure graphite mixed with active carbon from seaweed to reduce the use of graphite. The advantages of the Hummer method, which has been modified by [11], are that it is simple, production costs are lower, the materials used are environmentally friendly, the product produced is more orderly, and it takes a little less time when carrying out experiments.

Graphene oxide was synthesized using the modified Hummers method using H_2SO_4, KM_nO_4, H_2O_2, and N_aNO_3 compounds. The reason for using the manufacture of graphene oxide from a mixture of Graphite and seaweed is that Graphite is a natural rock that cannot be renewed, so its availability will decrease. Besides that, using Graphite will require high costs; therefore, the researchers conducted an experiment using seaweed mixed with Graphite, which can potentially be used as a layer of graphene oxide because seaweed contains active carbon.

This graphene oxide synthesis mixes Graphite with seaweed-activated carbon and is carried out with five composition variations. The result of this study is graphene oxide powder, which is then characterized to see crystal size, morphology, and optical properties. The modified Hummers method was chosen to synthesize graphene oxide because it has several advantages, such as lower production costs, the materials used are environmentally friendly, and the time needed is shorter. The products are produced more regularly [11].In this research, the author focuses more on the optical properties formed from the graphene oxide. They are making graphene oxide by varying the composition of the mixture between pure Graphite and seaweed, namely (100% graphite, 70% graphite-30% seaweed, 60% graphite-40% seaweed, 50% graphite-50% seaweed, and 100% grass sea). This composition variation is carried out so that each increase in the percentage of seaweed will affect the graphene oxide's optical properties and obtain optimum conditions. The optical properties of graphene oxide will be characterized using a UV-Vis Spectrometer [12].

This study aimed to see how the effect of adding seaweed to Graphite on the optical properties of the graphene oxide formed. The effect will be seen in every increase in the volume of seaweed carbon used. The optical properties observed include the level of absorbance or the ability of the material to absorb light, and the resulting Gap energy is also defined as the minimum energy required by electrons in the valence band to move to the conduction band (energy gap).

II. METHOD

Synthesis of graphene oxide from a mixture of seaweed charcoal with pure Graphite was carried out with five composition variations, namely 100% graphite, 70% graphite-30% seaweed, 60% graphite-40% seaweed, 50% graphite - 50% seaweed, and 100 % seaweed. The characterization results were obtained to see the effect of these composition variations on the optical properties of the resulting graphene oxide. This research is experimental, and the experiments were carried out in the materials physics laboratory, UNP chemistry laboratory, and the LLDIKTI Region X laboratory, Padang, West Sumatra. The main ingredients used to manufacture graphene oxide are pure Graphite and brown seaweed. These two materials will be mixed with several composition variations to obtain graphene oxide powder. Five composition variations were carried out, namely 100% graphite, 70% graphite - 30% seaweed, 60% graphite - 40% seaweed, 50% - 50%, and 100% seaweed. Graphene oxide was synthesized using a modified Hummers method, and this method uses acetic acid (H_2SO_4), potassium permanganate (KM_nO_4), hydrogen peroxide (H_2O_2), and distilled water. The results of this graphene oxide synthesis will be characterized using FTIR to see the functional groups formed, XRD to see crystal size, SEM to see morphology, and a UV-Vis spectrometer to characterize optical properties in the form of absorbance levels and energy gaps formed using the Tauc method. Plot. Tauc and colleagues first proposed the Tauc plot method to determine the band gap using absorbance data plotted with energy [13].

The first step in preparing brown seaweed was to take it from the coastal waters of Bungus, West Sumatera. We cleaned the seaweed and cut it into smaller pieces. Then, we dried it in the sun for two days. The next step was to dry the seaweed in an oven at 105℃ for 1 hour or 60 minutes to remove any remaining water content. You could refer to Figure 1(a) for pictures of seaweed after being washed and Figure 1(b) for pictures of seaweed after being baked.

(a) (b)

Fig 1. (a) samples of seaweed after washing, (b) samples of grass after baking.

Figure 1 (a) showed seaweed that had been washed clean, while Figure 1 (b) showed seaweed that had been baked. To obtain carbon, a sample was burned in a furnace. The burning process was carried out at a temperature of 300℃ for 30 minutes because this temperature produced the best quality carbon. Once burned, the samples were crushed and filtered through a 170-mesh sieve to obtain fine carbon. The seaweed carbon was then activated using KOH compounds to produce activated carbon. To do this, 8 grams of seaweed carbon was mixed with a solution of 8 grams of KOH dissolved in 100 ml of distilled water. The mixture was stirred for 15 minutes and left to stand for 24 hours. After that, the solution was neutralized by washing it with distilled water, and the sample was baked in an oven at 105℃ for 3 hours.

III. RESULTS AND DISCUSSION

Figure 2 shows images of seaweed samples: (a) after being furnaced, (b) mashed, and (c) carbon after activation using KOH

(a) (b) (c)

Fig 2. (a) Seaweed samples after being furnaced, (b) Furnanced seaweed samples which were mashed, (c) Seaweed carbon after it was activated (activated carbon).

Figure 2(a) shows a sample of seaweed treated with carbon in a furnace. In Figure 2(b), the carbon is crushed using a mortar and pestle, and in Figure 2(c), the resulting carbon is activated using KOH.

The second step involves synthesizing graphene oxide. To do this, a mixture of graphite and activated seaweed carbon is weighed in a 1.5-gram ratio. 34.5 ml of H_2SO_4 and 0.75 ml of N_aNO_3 are added to the weighed sample and then stirred for 20 minutes. The sample is then treated with ice cubes for two hours to lower the risk of explosion when KM_nO_4 is added. Afterward, 4.5 grams of KM_nO_4 are gradually added to the sample, which is stirred for 30 minutes without ice cube treatment.

The material is stirred in a fume cupboard while adding 69 ml of distilled water slowly. It is then stirred for 20 minutes before adding another 100 ml of distilled water. The solution is mixed with 1.5 ml of

peroxide acid (H_2O_2) to reduce the bubbles resulting from mixing KM_nO_4, and then another 50 ml of distilled water is added.

The sample is stirred and sonicated for two hours using ultrasonic to flake the graphite oxide into graphene oxide. It is then left to stand for one day to form solid and liquid phases. Distilled water is repeatedly replaced until a neutral pH of 7 is obtained, and then the sample is oven-dried at 60°C for 12 hours. The resulting product is graphene oxide powder, characterized using FTIR, XRD, SEM, and UV-Vis Spectrometer.

Graphene oxide is a material that consists of several functional groups. The FTIR tool is used to characterize graphene oxide and identify the composition of the functional groups formed. The test is carried out from a wavelength of 500-4000 cm-1. The results of the characterization of graphene oxide with FTIR are presented in a graph showing the peaks containing functional groups. The graph of the results of the FTIR characterization of the five graphene oxide samples can be seen in Figure 3.

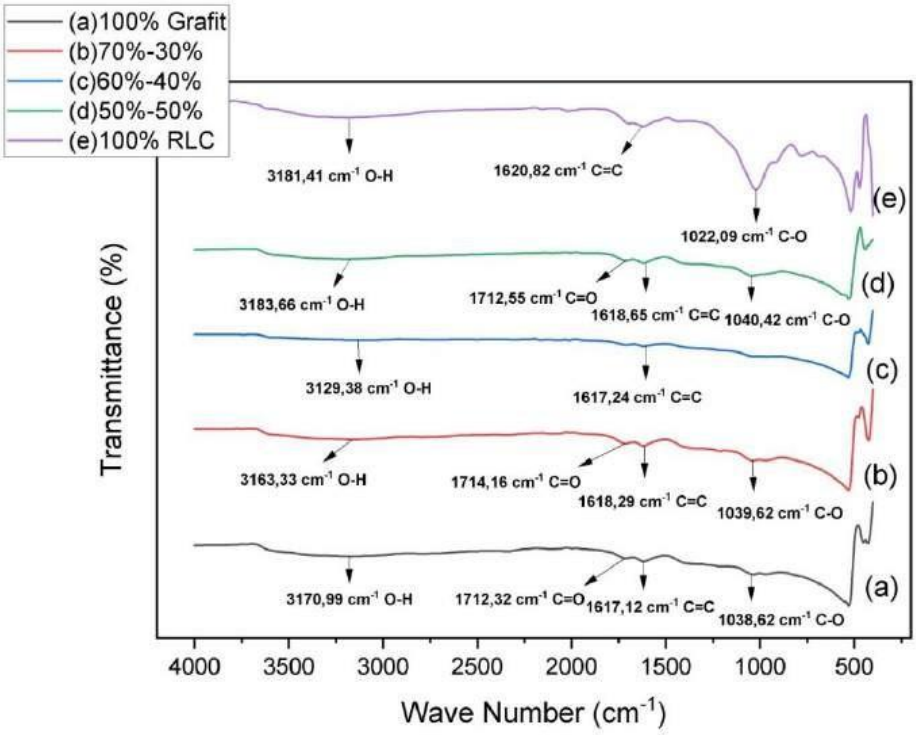


Fig 3. FTIR test results of graphene oxide samples for five composition variations

From Figure (3), it can be seen that the functional groups detected from five graphene oxide samples based on variations in composition, namely the presence of carbon (C), Hydrogen (H), and Oxygen (O) bonds which form functional group bonds O-H, C=O, C =C, and C-O.

This shows that graphene oxide has been formed, where graphene oxide contains carbon (C), hydrogen (H), and oxygen (O) bonds. The O-H functional group in graphene oxide indicates the presence of carboxylic acid compounds, alcohol, phenol (H bonds), and phenol (monometer). The C=O functional group bonds in graphene oxide indicate the presence of aldehyde, ketone, carboxylic acid, and ester compounds. C=C functional group bonds in graphene oxide, which indicate the presence of alkene compounds and aromatic rings. C-O functional group bonds in graphene oxide indicate the presence of ester, alkyl, vinyl ether, and aromatic compounds.

The characterization results of graphene oxide for five composition variations provide information in graphs displaying the diffraction peaks. The graph of the results of the XRD characterization of the five graphene oxide samples can be seen in the following figure (4):

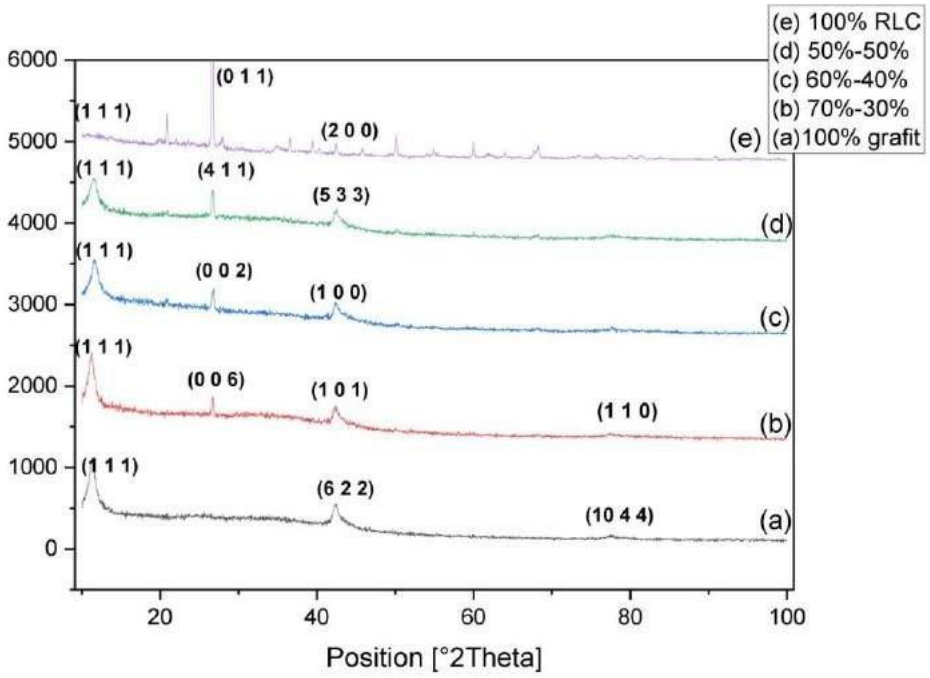
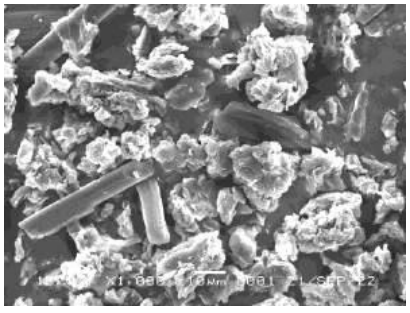


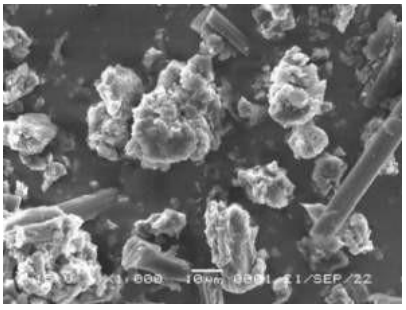
Fig 4. XRD pattern of graphene oxide samples for five composition variations

The graph in Figure 4 displays the diffraction peaks and their corresponding Miller indices. This data can be used to determine the average size of the graphene oxide crystals. In analyzing the XRD data processing results, it is evident from the graph that a composition of 100% graphite exhibits no discernible peak at an angle of 24 degrees. However, with the addition of seaweed, visible peaks emerge at this angle for compositions of 70% graphite - 30% seaweed, 60% graphite - 40% seaweed, 50% graphite - 50% seaweed, and 100% seaweed. This observation suggests that incorporating seaweed enhances the quality of produced graphene oxide. Furthermore, correlating these findings with the energy gap results reveals a trend: as the proportion of seaweed increases, the energy gap decreases, approaching the ideal energy gap value. Thus, it can be inferred that higher seaweed compositions yield graphene oxide with superior optical properties.

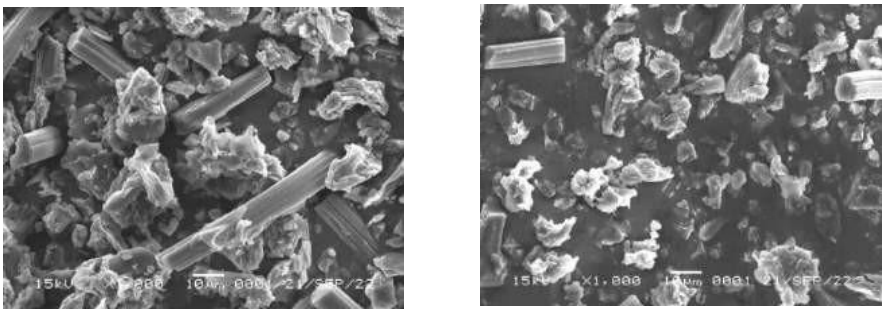
The characterization results using SEM provide information in the form of morphology and particle size of the resulting graphene oxide. The morphological forms of the five graphene oxide samples can be seen in Figure 5 (a-e) :



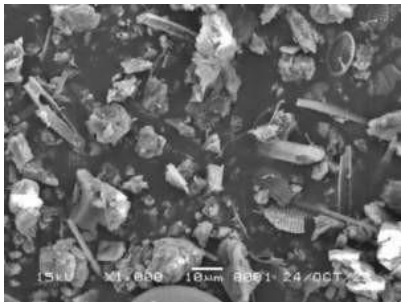
(a)100% Graphite



(b) 70% Graphite – 30% seaweed carbon



(c) 60% Graphite - 40% seaweed carbon (d) 50% - 50%



(e) 100% seaweed carbon

Fig 5. The morphology of the SEM test results (a) 100% graphite, (b) 70% graphite-30% seaweed carbon, (c) 60% graphite-40% seaweed carbon, (d) 50%-50%, (e) 100% seaweed carbon

From Figure 5(a)-5(e), it can be seen that the morphology of graphene oxide is that some are in the form of lumps, some are hollow layers, and some are flaky, which indicates that graphene oxide has been successfully formed. From the data obtained, the average grain size of each sample can be found.

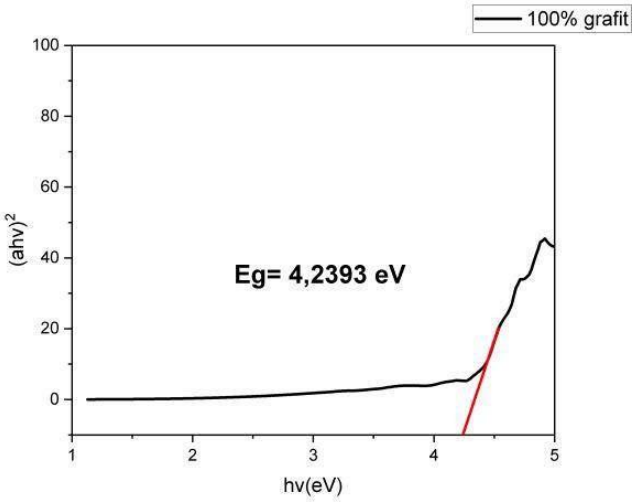
The characterization results used a UV-Vis spectrometer to determine the optical value of the resulting graphene oxide. The data obtained is then processed, and the absorbance level of each sample is obtained. The absorbance level of the five graphene oxide samples can be seen in the table (1) below:

Table 1. UV-Vis Spectrum and Absorbance

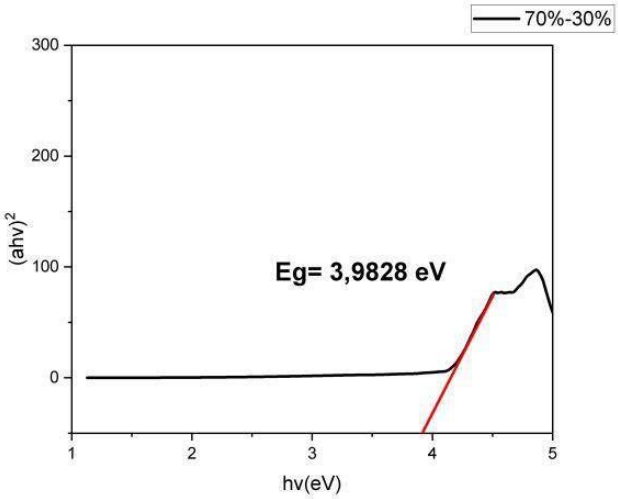
Composition	Wavelength (nm)	Absorbance
100% Graphite	203	30,971
70% graphite – 30% seaweed carbon	275	42,55
60% graphite – 40% seaweed carbon	272	32,754
50% - 50%	245	49,547
100% seaweed carbon	294	17,991

Table (1) shows the highest absorbance values obtained at specific wavelengths, and the data shows that the obtained absorbance values are still in the graphene oxide wavelength range. The ideal absorbance peak is around a wavelength value of 250 nm [14]. There is also a study on graphene oxide, which gets an absorbance peak at a wavelength of 230 nm, and he says this is included in the absorbance range of graphene oxide [15]. A study also uses the LPE method to get an absorbance peak of 270 nm [16].

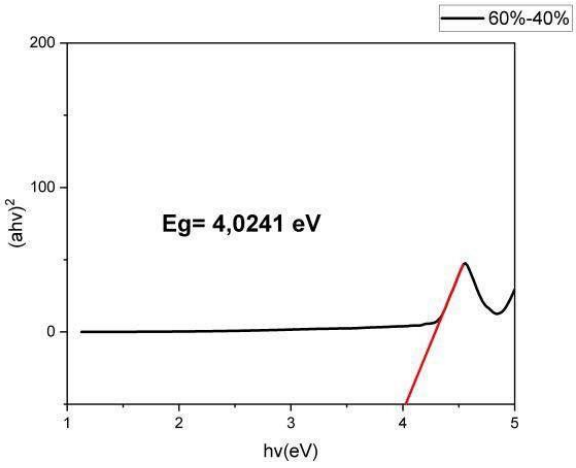
The optical value of a graphene oxide is also determined by the value of the energy gap/energy band gap obtained from the data processing results using the Tauc Plot method. The following is a graph of the results of data processing that displays the gap energy for each sample, as shown in Figure 6 :



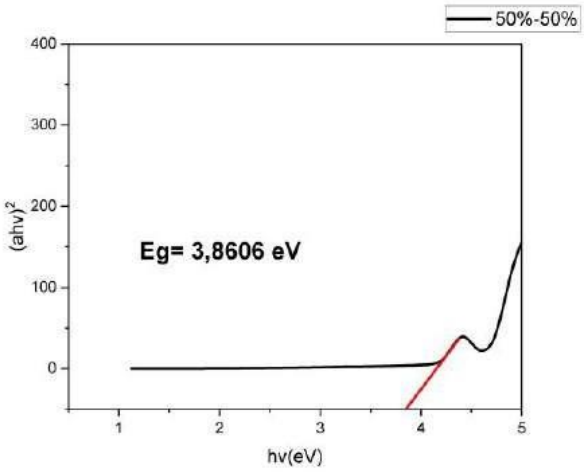
(a) 100% Graphite



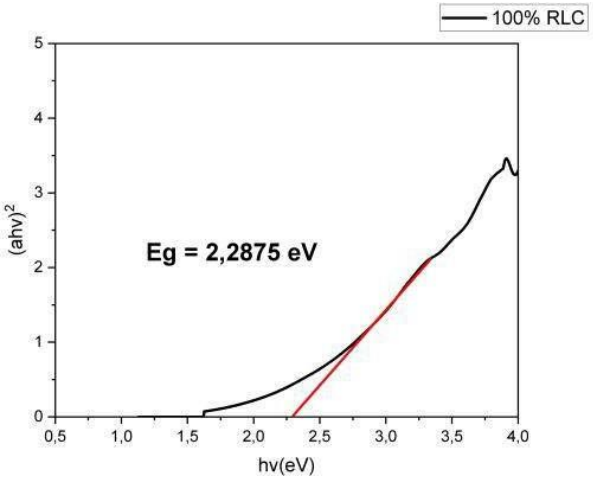
(b) 70% Graphite- 30% seaweed carbon



(c) 60% Graphite- 40% seaweed carbon



(d) 50% Graphite – 50% Seaweed carbon



(e) 100% seaweed carbon

Fig 6. Gap energy values (a) 100% graphite, (b) 70% graphite-30% seaweed carbon. (c) 60% Graphite- 40% seaweed carbon, (d) 50%-50%, (e) 100% seaweed carbon

Based on the graphs in Figure 6(a) – 6(e), the band gap energy values from lowest to highest sequentially are in the composition of 100% seaweed with 2.2827 eV, 50% - 50% with 3.8606 eV, 70 % graphite – 30% seaweed with 3.9828 eV, 60% graphite – 40% seaweed with 4.0241 eV, and 100% graphite with 4.2392 eV. Graphene oxide generally has a band gap value of around 2.7 – 4.2 eV [14]. For example, research on graphene oxide from graphite powder produces a band gap value of around 4.2 eV [16], and some obtain band gap values in the 2.9 - 4.4 eV [17]. From the results of band gap data processing using the Tauc Plot method, band gap results are obtained in the range of 2.2 eV – 4.2 eV. This data is under the graphene oxide band gap obtained by other researchers, proving that the sample made has formed graphene oxide. / synthesis of graphene oxide from pure graphite and seaweed was successfully carried out. Judging from the band gap data obtained, the more seaweed composition is used, the more the band gap value of graphene oxide will be reduced. However, the band gap value obtained is still in the semiconductor band gap range, below 6 eV, which means that the graphene oxide powder made can be utilized. as a semiconductor device. As for the graphene material itself, it does not have a band gap or a value of 0 band gap [16]. This graphene oxide will be applied as a semiconductor device in which the semiconductor has a band gap of <6 [13].

IV. CONCLUSION

According to research conducted on the effects of adding Seaweed Charcoal (Sargassum sp) with pure Graphite on the optical properties of graphene oxide synthesized using the modified Hummer method, it was found that the absorbance values of each sample were within the range of 200 nm to 300 nm, which is the range of absorbance values of graphene oxide. The results of the energy gap analysis indicate that the 100%

graphite variation has the highest gap value, while the 100% seaweed variation has the lowest gap value. This proves that adding seaweed to pure Graphite reduces the energy gap of graphene oxide.

For future research, it is recommended to pay attention to several material conditions and use samples with high carbon content while using different variations, to provide better results..

ACKNOWLEDGMENT

The author thanks the heads of UNP material physics and chemistry labs, and LLDIKTI Region X Padang lab for support during research.

REFERENCES

[1] W. F. Caspary, J. Schäffer, G. Brunner, G. Schmidt, and W. Creutzfeldt, "14C-aminopyrine-(pyramidone) respiratory test--a new quantitative liver function test," *Verh. Dtsch. Ges. Inn. Med.*, vol. 82 Pt 1, pp. 286–289, 1976, doi: 10.1007/978-3-642-85451-4_42.

[2] A. S. Suparmi, "Kajian Pemanfaatan Sumber Daya Rumput Laut Dari Aspek Industri dan Kesehatan," *J. Maj. Ilm. Sultan Agung*, vol. 44, no. 118, pp. 95–116, 2013.

[3] ali rido Sri Sedjati, Suryono, Adi sentosa, Endang supriyantini, "Aktivitas Antioksidan dan Kandungan Senyawa Fenolik Makroalga," vol. 20, no. November, pp. 117–123, 2017.

[4] I. Kusumaningrum, R. B. Hastuti, and S. Haryanti, "Pengaruh Perasan Sargassum crassifolium dengan Konsentrasi yang Berbeda terhadap Pertumbuhan Tanaman Kedelai (Glycine max (L) Merill)," *Bul. Anat. dan Fisiol.*, vol. XV, no. 2, pp. 17–13, 2007.

[5] S. N. Lim, P. C. K. Cheung, V. E. C. Ooi, and P. O. Ang, "Evaluation of antioxidative activity of extracts from a brown seaweed, Sargassum siliquastrum," *J. Agric. Food Chem.*, vol. 50, no. 13, pp. 3862–3866, 2002, doi: 10.1021/jf020096b.

[6] H. Samee, Z. X. Li, H. Lin, J. Khalid, and Y. C. Guo, "Anti-allergic effects of ethanol extracts from brown seaweeds," *J. Zhejiang Univ. Sci. B*, vol. 10, no. 2, pp. 147–153, 2009, doi: 10.1631/jzus.B0820185.

[7] D. Saputra Lega Wira Made,Ariani Panti Risa, "Cookies Berkadar Serat Tinggi Substitusi Tepung Ampas Rumput Laut Dari Pengolahan Agar-Agar Kertas," *Bosaparis Pendidik. Kesejaht. Kel.*, vol. 10, no. 1, pp. 49–60, 2019, [Online]. Available: https://www.google.com/url?sa=t&rct=j&q=&esrc=s&source=web&cd=&ved=2ahUKEwjPj-Cvldb0AhX0UWwGHTE7BSgQFnoECAkQAQ&url=https%3A%2F%2Fejournal.undiksha.ac.id%2Findex.php%2FJJPKK%2Farticle%2Fdownload%2F22158%2F13778&usg=AOvVaw1czbfS6A_yRr7NWMqSpL4v

[8] S. C. Ray, *Application and Uses of Graphene*. Elsevier Inc., 2015. doi: 10.1016/b978-0-323-37521-4.00001-7.

[9] N. U. R. Anggita and Dewayanti, "Synthesis of Graphene-Like Based on Coconut Shell Biomass and Its Applications As Electrodes in Metal- Air Battery," 2018.

[10] F. Azka, "SINTESIS DAN KARAKTERISASI GRAPHENE OXIDE TERKOMBINASI NANOPARTIKEL PERAK DALAM FASE CAIR," *Plant, Cell Environ.*, vol. 29, no. 2, pp. 192–201, 2006, doi: 10.1111/j.1365-3040.2005.01412.x.

[11] D. C. Marcano *et al.*, "Improved synthesis of graphene oxide," *ACS Nano*, vol. 4, no. 8, pp. 4806–4814, 2010, doi: 10.1021/nn1006368.

[12] I. R. Solo, M. Bukit, A. Z. Johannes, and R. K. Pingak, "Kajian Awal Sifat Optik Graphene Oxide Berbahan Dasar Arang Sekam Padi Dengan Metode Liquid Phase Exfoliation Menggunakan Alat Bantu Blender Dan Ultrasonic Cleaner," *J. Fis. Fis. Sains dan Apl.*, vol. 5, no. 2, pp. 145–148, 2020, [Online]. Available: https://ejurnal.undana.ac.id/FISA/article/view/3472/2289

[13] Y. Oktaviani and Astuti, "Sintesis Lapisan Tipis Semikonduktor dengan Bahan Dasar Tembaga (Cu) Menggunakan Chemical Bath Deposition," *J. Fis. Unand*, vol. 3, no. 1, pp. 53–58, 2014.

[14] A. Redinger and S. Siebentritt, "Loss Mechanisms in Kesterite Solar Cells," *Copp. Zinc Tin Sulfide-Based Thin-Film Sol. Cells*, vol. 627, pp. 363–386, 2015, doi: 10.1002/9781118437865.ch16.

[15] D. Bhatnagar, S. Singh, S. Yadav, A. Kumar, and I. Kaur, "Experimental and theoretical investigation of relative optical band gaps in graphene generations," *Mater. Res. Express*, vol. 4, no. 1, 2017, doi: 10.1088/2053-1591/4/1/015101.

[16] A. K. Geim and K. S. Novoselov, "The rise of graphene," *Nanosci. Technol. A Collect. Rev. from Nat. Journals*, pp. 11–19, 2009, doi: 10.1142/9789814287005_0002.

[17] H. C. Hsu *et al.*, "Graphene oxide as a promising photocatalyst for CO2 to methanol conversion," *Nanoscale*, vol. 5, no. 1, pp. 262–268, 2013, doi: 10.1039/c2nr31718d.