SYNTHESIS OF REDUCED GRAPHENE OXIDE AND ZINC OXIDE COMPOSITE FROM CANDLENUT SHELL CHARCOAL (*Aleuritas moluccana*)

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Abstract: Candlenut shell is one of the by-products of the hazelnut processing industry which has not been widely utilized. However, the carbon content from the candlenut shell can be used as charcoal, which has the potential as the base material for making reduced graphene oxide (GO) capacitance products. After the composite process, characterization of the rGO and its composites was carried out with FTIR and XRD instruments. The synthesis results obtained are combined with ZnO to determine the value of the electrical result. From FTIR characterization, it was found that C=C, C-O, and O-H functional groups were formed, and the XRD measurement resulted in a peak diffractogram at 2θ ~ 23,96°, which is typical for rGO material. The result of testing with a conductivity meter at a rGO-ZnO 0:1 ratio, produced a capacitance value of 4,72 mF, in terms of 1:2, 6,34 mF; 1:1, 7,36 mF; 2:1, 5,18 mF; and 1:0, 4,28 mF. The optimum ratio of rGO-ZnO with the highest capacitance value was found, which is the ratio of 1:1 with a capacitance value of 7,36 mF.
Keywords: Graphene oxide; Capacitance; Composite; Candlenut shell

Abstrak: Tempurung kemiri merupakan salah satu hasil samping dari industri pengolahan kemiri yang masih belum banyak dimanfaatkan secara luas. Akan tetapi kandungan karbon pada tempurung kemiri dapat dijadikan arang yang berpotensi sebagai salah satu bahan dasar pembuatan Grafena Oksida(GO) tereduksi. Hasil sintesis yang didapatkan dikompositkan dengan ZnO untuk mengetahui nilai kapasitansi yang dihasilkan. Setelah proses komposit, dilakukan karakterisasi terhadap rGO dan kompositnya dengan instrumen FTIR dan XRD kemudian diuji nilai kapasitansi elektrik yang dihasilkan dengan alat multimeter. Berdasarkan hasil karakterisasi FTIR, didapati terbentuk gugus C=C, C-O, O-H dan uji XRD menghasilkan nilai 2θ ~ 23,96^{o} yang mengindikasikan rGO .Hasil pengujian dengan konduktimeter menghasilkan data perbandingan rGO-ZnO 0:1 menghasilkan nilai kapasitansi 4,72 mF, 1:2, 6,34 mF; 1:1, 7,36 mF; 2:1, 5,18 mF; dan 1:0, 4,28 mF. Diperoleh perbandingan optimum rGO-ZnO dengan nilai kapasitansi paling besar yaitu perbandingan 1:1 sebesar 7,36 mF.
Kata kunci: Grafena Oksida; Kapasitansi; Komposit; Tempurung Kemiri

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Introduction

The coconut shell partially consists of lignin, cellulose, and hemicellulose (Liyanage & Pieris, 2015) and 57.11% carbon, 42.67% oxygen, and 0.23% other materials (Hidayat et al., 2018). The most optimal results were obtained on graphene with the smallest particle size and a synthesis time of 5 hours (Vinanza, 2018). Then it was used as a supercapacitor with the largest specific energy capacity and a medium discharge time (Zheng et al., 2017).

Technological developments in the modern era in the field of materials range from the discovery of plastics and synthetic fibers to composite materials. Graphene has unique characteristics compared to other materials, such as a large surface area, high electrical conductivity, and good optical and thermal properties. Graphene material is stronger and more durable because it has a hexagonal compound structure with a strength of 42 N/m (Marcelina et al., 2017). Graphite oxide becomes thinner and reduces the number of sheets and produces compounds graphene oxide. Graphene oxide is what is then reduced chemically to produce rGO (Lee et al., 2017). Making rGO can be done by reducing GO material with ammonium thiocyanate as an oxygen-reducing agent in its structure. (Li et al., 2017). Graphene applies graphene apply fields, such as batteries, sensors, and a supercapacitor (Tiwari et al., 2017). Graphene has the ability to carry an electric charge of up to 200,000 cm^2/Vs. The energy difference of the rGO shell is still around 2 electron volts (Mas'udah et al., 2016). The problem now is that it is still hard to use graphene's better properties on a large scale with the way things are made now.

The graphene production process can use various methods. For example, the CVD (Chemical vapor deposite) method can produce high-quality graphene with high purity. Another method is high-quality graphene with high purity. Another method, peeling, produces graphene in large quantities and at low cost but has lower purity, and the resulting graphene is not continuous (sheets) but in the form of powder or flakes (Tang et al., 2017). Another method is to peel layers of graphite or carbon crystals down to the micrometer scale using tape (Gerasimov, 2017). Therefore, top-down approaches, such as chemical peels, are more often used in industry. In addition, another advantage of the top-down method is that it can use carbon sources.

The use of candlenut shell as a raw material for producing graphene can increase its benefits and economic value. The resulting graphene will be composited with zinc oxide to become a capacitor to test its capacitance value.

Methods

Materials and Instruments

The materials used are candlenut shell, sulfur acid (H_2SO_4 95-98%, Merck), potassium permanganate ($KMnO_4$ 97%, Merck), peroxide (H_2O_2 30%, Merck), hydrochloric acid (HCl 37%, Merck), ethanol absolute, butanol and demineralised

water. Instruments used for characterization are X-Ray Diffraction (XRD, Shimadzu-7000) and IR Spectroscopy (Thermo Scientific Nicolat iS10).

Sample Preparation

1 kg of candlenut shells is burned using a carbonization tank. The shells that have been converted into charcoal are then mashed using a blender and filtered through a 230 mesh sieve to produce the fine charcoal powder. The charcoal is then made to work physically by putting it in an oven at 60 °C for 6 hours.

Oxidation Stage

24 g of candlenut shell charcoal powder was added to a beaker containing 560 mL of concentrated sulfuric acid. The beaker was stored in an ice bath, then 20 g of $KMnO_4$ was added little by little so that the temperature of the mixture did not exceed 20 °C, and then 1800 mL of distilled water was added. The suspension was then stirred for two hours at 35 °C. The suspension was allowed to stand (oxidation process) for 5 days.

Reduction Stage

After the oxidation process is complete, add 2000 mL of water and 60 mL of H_2O_2 (reduction process), and this process causes the solution to foam and the temperature of the mixture to increase. During the reaction, the color of the mixture will change from dark brown to yellow. The mixture was then washed with 1 L of HCl solution (1:10 ~ HCl : water) using filter paper so that metal ions could be removed. At this stage, a paste will be produced, which is then dried at 60 °C for 6.5 hours until a solid is formed. After that, the solid is placed in 2000 mL of water and allowed to stand for 3 hours. Purification is done by washing the suspension with a large amount of water.

Ultrasonic Exfoliation

The paste that had been collected from the filter paper was mixed into 300 mL of deionized water and then sonicated with a sonicator for 20 minutes. The result of the brown dispersion of reduced graphene oxide (rGO) was then filtered using a vacuum filter so that the water and residue separated. This residue was then scraped off the filter paper and allowed to dry. Furthermore, dry rGO will be obtained by the dehydration process. The dehydration process was started by heating the rGO in a dish at 90°C for 1 hour. The rGO flakes will be obtained by scraping the rGO solids from the cup.

Composite Manufacturing

Both materials, namely rGO and ZnO, were prepared. In each variation, they were put in a beaker glass and given a solution of 1 butanol as a mixing solution for the two ingredients. The mixing of these two materials was carried out for five hours using a magnetic stirrer. The two ingredients that have been mixed will then be heated using an oven for ± 5 hours.

Result and Discussions

Old coconut shells that have undergone a carbonization process at 400 ^{o}C have the main molecular bonds of graphene, namely C = C and C-C. It also contains many other bonds such as C-H, C-O, C=O, and O-H, indicating the presence of the rGO phase (Nugraheni et al., 2015). The results of the spectroscopic test showed peaks at 1300 cm^{-1} and 1590 cm^{-1} with an ID/IG ratio of about 2.5, and the XRD test results showed peaks at 250 and 400, which indicated the presence of the rGO phase (Prasetya et al., 2015).

Graphene production is mostly done by oxidizing graphite to graphene oxide (Harahap, 2018) and using the Hummer method of graphene oxide (GO), but the presence of oxygen in graphene oxide reduces GO performance. Therefore, it is necessary to carry out a reduction process to remove oxygen from GO so that it leaves the graphene layer (Lasmana et al., 2016).

Pecan skin is selected dry so that it is easier to turn into charcoal. The charcoal that has been obtained is crushed and filtered using a sieve shaker with a sieve size of 230 mesh. Fine charcoal was dissolved in an HCl solution and washed with distilled water.

The next process after going through the sample preparation process is the synthesis of graphene oxide, which is carried out in three stages, namely oxidation, reduction, and ultrasonication. During oxidation, the candlenut shell charcoal sample was reacted with sulfuric acid (H_2SO_4) and potassium permanganate ($KMnO_4$). Sulfuric acid (H_2SO_4) so that oxidation can occur and potassium permanganate ($KMnO_4$). This process is carried out in an ice bath and kept below 20 ^{o}C. Then add 300 mL of distilled water and let it stand for 5 days.
The reaction that occurs during oxidation causes some dark purplish color changes due to the original color of potassium permanganate, then changes to a greenish color. Overall, the oxidation process of charcoal samples aims to introduce oxygen, hydroxyl, and ketone groups into the crystal structure of carbon in order to create distance between layers and facilitate the separation process into the rGO layer.

Reduction aims to remove or reduce the functional groups formed in the oxidation process to obtain rGO. Reduction is carried out with hydrogen peroxide (H_2O_2) and distilled water. The reduction process is characterized by a change in the color of the sample solution from dark brown to dilute yellow, accompanied by foam and smoke. This process is also carried out in an ice bath to avoid the occurrence of too high a temperature due to an exothermic reaction. After the reduction process is complete, washing with hydrochloric acid (HCl) is carried out to clean the remaining metal ions, and finally, the solution is diluted with distilled water so that the pH of the solution is close to neutral.

FTIR Characterization Result

The resulting rGO was composited with ZnO in four comparison variations. ZnO was chosen as a composite material because it has a wide band gap, high

electron mobility, and good transparency, as well as high heat capacity and conductivity. From some of these variations, the characteristics will be compared through the FTIR, XRD, and electrical capacitance tests.

Sharp peaks indicate that in GO there is an oxygen functional group in its structure. The rGO shows a wide peak diffractogram at a value of 2θ = 24.97° (hkl 002) and a small peak at a value of 2θ = 42.42° (hkl 100).

Table 1. FTIR characterization result

rGO	rGO 1 g – ZnO 0,5 g	rGO 0,75 g – ZnO 0,75 g	rGO 0,5 g – ZnO 1 g	Functional Groups rGO	Wave Number (cm^{-1})
3359,41	3232,31	3233,17	3366,07	O–H	3550 – 3200
1593,63	1577,98	1575,67	1577,25	C=C	1900 – 1500
-	-	-	-	C=O	1740 – 1720
1193,03	1189,18	1192,24	1193,15	C–O	1260 – 1000

Each functional group that forms in the sample absorbs infrared light differently. This lets you know that the desired compound has formed.

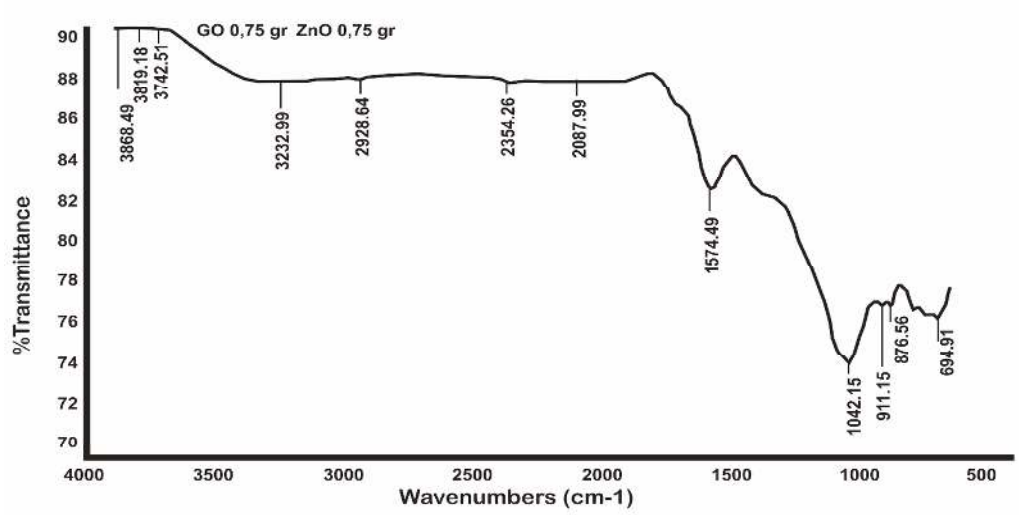

Figure 1. FTIR graphic of rGO

From the results of the tests carried out, the rGO sample was detected to contain the OH functional group as indicated by the absorption at a wavelength of 3359.41 cm^{-1}, the C=C group at a wavelength of 1593.63 cm^{-1}, and the CO group at a wavelength of 1193.03 cm^{-1}. According to Hidayat et al. (2018), the oxidation process that occurs is going quite well. The oxidation process of carbon to graphene oxide causes the formation of oxygen-containing functional groups such

as O-H, C-O, and C=O. While the reduction process that occurs is imperfect, according to research conducted by Husnah et al. (2015), the process of reducing graphene oxide (GO) to reduced graphene oxide (rGO). The two O-H. groups which appear in the GO and rGO (M.khan et al., 2020) spectra are hydroxyl and carboxyl groups (Hidayat et al., 2018), Even though the reduction process is not perfect, the reduction process still continues, as indicated by the undetection of the C = O group and the formation of the C=C group, which is the main constituent of the graphene layer structure.

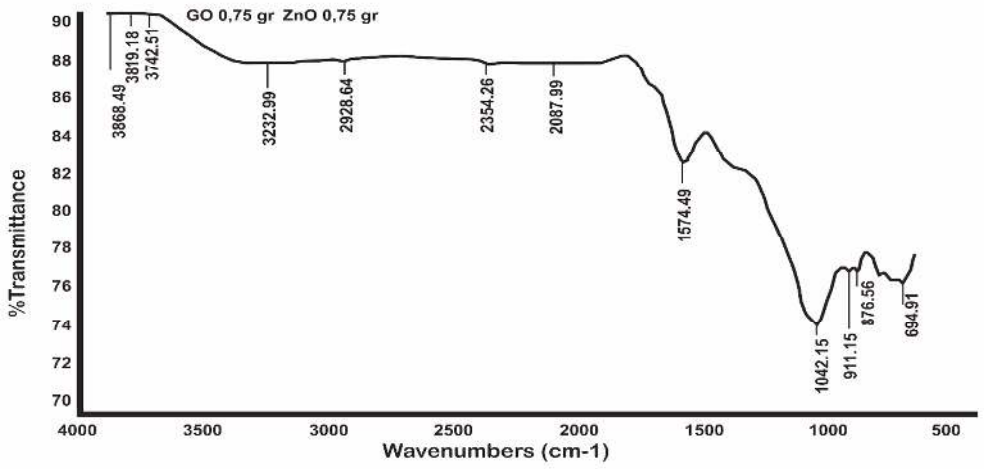

Figure 2. FTIR graphic of rGO-ZnO

The absorption of C–OH and C=O in the rGO material is no longer visible. This indicates that the sample has not exfoliated well even though the C = C peak is higher than that of the graphite sample (Alam et al., 2017). This also indicates that rGO has been formed with the reduction of the OH group and the loss of the C=O group.

FTIR test results of the four variations of the rGO composites showed that the results did not differ greatly between each sample. Each sample has functional groups O–H, C=C, and C–O, which indicates that the composite process runs well without changing the chemical properties of rGO.

XRD Characterization Result

The next sample characterization was carried out using the X-Ray Diffraction (XRD) tool. In figure 3, it can be seen that a peak formed at 2q 23.90^{o} and d spacing of 3.72021 A, which states that the peak or peak of rGO is at 2q 23 – 24^{o}. The results of the characteristic spectrum of the rGO peak obtained are in accordance with the study (M.Husnah et al., 2017). However, based on Figure 3, in addition to the diffraction peaks that indicate the presence of rGO, there are also other peaks found in the study. 1 phase of rGO has a band at an angle of 2θ by 25 ° (Setiadji et al., 2018).

This shows that there are still impurities in the sample, possibly as a result of the reduction process which is imperfect.

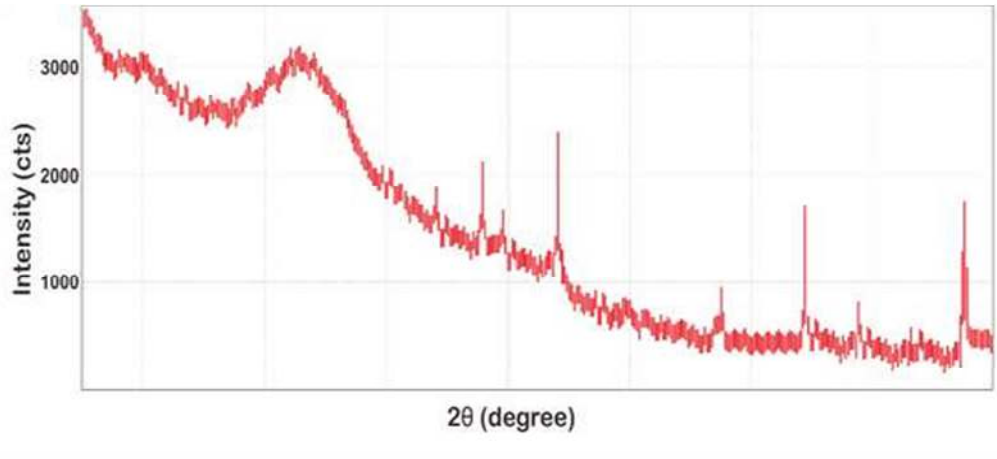

Figure 3. XRD diffraction pattern of rGO

In figure 3, it can be seen that the reduced phase of graphene oxide formed is an amorphous phase, which is characterized by a wide and quite sloping peak shape, in contrast to the crystal phase, which generally has sharp and narrow peaks. The thing that causes the rGO produced in this research to have an amorphous phase is the synthesis method used, namely the top-down method, which generally produces powder and not a wide sheet, so that the X-rays that are emitted are not diffracted at one angle just like crystals, but are diffracted in several adjacent corners to form a sloppy pattern.

In the rGO and ZnO composite, a sample with a 1: 1 ratio was used. The resulting diffraction pattern can be seen in figure 4, which shows the rGO pattern at an angle of 2q 23.96^{o}. The addition of ZnO caused a decrease in the intensity of rGO, seen in a combination of diffraction patterns between rGO and ZnO, but there was a change in intensity at several peaks. But in general, neither the structure nor the distance between the layers of reduced graphene oxide changed importantly.

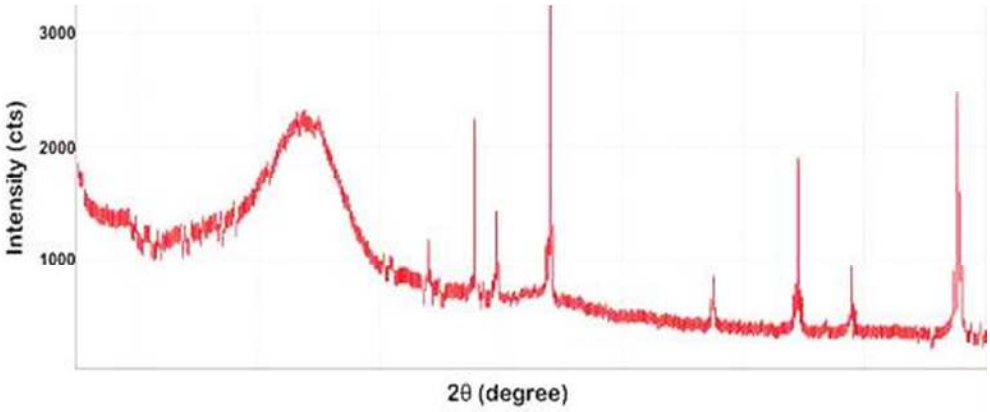

Figure 4. XRD diffraction pattern of rGO-ZnO

This indicates that there has been a physical mixing between rGO and ZnO (composite) without causing a chemical change to rGO. The chemical reduction of GO to rGO was shown from the expansion and shift of the XRD peak to be about 2θ=25° with an interlayer distance of 3.49Å. Previous research conducted (Li et al., 2017) stated that graphite and rGO have a 2θ band at 25-2°, while graphene oxide has a 2θ band at 1-12° and there is a diffraction peak (002) at 2θ ~24.04˚ which indicates the distance between layers in rGO and a diffraction peak (101) at 2θ ~44.02˚, while at 2θ = 9.8˚-11.5 (Puspitasari, 2017).

Capacitance Test Result

Electrical capacitance is one of the important electrical properties of a material as well as its electrical conductivity. The electrical conductivity of a material determines how energy can be transferred and distributed efficiently and safely. While electrical capacitance is the ability of a series of systems to store energy, as conductivity affects modern life, electrical capacitance also determines how energy can be stored and used effectively. Its application can be seen in supercapacitors, which have a very large potential role for energy needs in the future.

The rGO and ZnO capacitance testing is done by assembling it into a simple capacitor. In the making of capacitors, PVA glue is used as a binder, PVA glue is used because it is soluble in water and does not interfere too much with the capacitance properties. As an electrolyte, sodium sulfate is used because it is cheaper and safer than sulfuric acid, but it still works well. In the results of the tests carried out, the capacitance values of pure rGO and pure ZnO showed the lowest values, namely 4.28 mF and 4.72 mF. While the capacitance ratio of rGO : ZnO 1:2 and 2:1 increased to 5.18 mF and 6.34 mF, the highest capacitance value was obtained at concentrations of rGO and ZnO 1:1, 7.36 mF which stated that a good mixture of ZnO and composites rGO will increase the capacitance value of the material. That the rGO-ZnO composite could be made better so that it can be used to make supercapacitors. Performance of supercapacitor cells in previous

studies with specific capacitance values ranging from 6.5 to 27.3 mF/cm^2 (Fitrilawati et al, 2019), (B.Wei et al, 2018).

Figure 5. Graphic of capacitance test

The regression value of the research data is 0.037. The regression results are close to zero, indicating that the addition of ZnO to rGO in the composite and vice versa, has no linear effect on the resulting electrical capacitance value. Or in other words, the rGO - ZnO ratio variable has no significant effect on the electrical capacitance variable. However, from the data obtained, it can be seen that the optimum composition with the largest electrical capacitance value is the ratio of rGO - ZnO one to one with a capacitance value of 7.36 mF.

Conclusion

The reduced graphene oxide can be identified from the FTIR characterization, which shows the presence of a C=C group, and the XRD characterization, which shows a peak of 2q 23.90^o. Variations in the ratio of rGO-ZnO were tested at different capacitance values. The 0:1 rGO-ZnO ratio yielded 4.72 mF; 1:2, 6.34 mF; 1:1, 7.36 mF; 2:1, 5.18 mF; and 1:0, 4.28 mF. The highest capacitance value is a ratio of 1:1 with a capacitance value of 7.36 mF.

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