Synthesis and characterization of graphene oxide as dielectric material for parallel plate capacitor

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Graphite acts as an electrical conductor due to the delocalization of electrons between the surfaces. Here, graphene oxide (GO) is produced from graphite via the Hummer's method. Furthermore, we have produced simple parallel plate capacitors containing the synthesized GO as the dielectric material with variation of the GO mass, *i.e.*: 0.0 g; 0.2 g; and 0.8 g. Moreover, we determine the capacitance values obtained from the capacitors using a multitester device. Characterizations of the GO material are conducted using X-ray diffraction (XRD), ultraviolet-visible (UV-Vis) spectrophotometer, and Fourier transform infrared (FTIR) spectroscopy. The result of the UV-Vis test shows a shouldering peak at a wavelength of 300 nm indicating the existence of oxygen functional groups of the GO sample. The XRD test shows the existence of GO material with 2θ at 12° and an indication of a transition from GO to rGO in an amorphous phase. Finally, the FTIR test produces transmittance bands with functional groups of C=C, CO_2, CO, and O-H. The results show that the capacitance values of the capacitors using GO as the dielectric material are higher than without using the GO sample. The highest capacitance value is obtained for the capacitor with 0.2 g of GO, *i.e.*: 165.02 μF.

Keywords: GO; Dielectric material; Parallel plate capacitor.

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1. Introduction

Today's electronic technology is growing rapidly. This rapid growth is evidenced by the need for energy storage that continues to be developed. Capacitor is one of the electronic circuits that can be used for energy storage. Traditionally, capacitors can be grouped into ceramics, polymers, and ceramic-polymer composites capacitors. Capacitors consist of dielectric materials, which can increase the capacitance of the capacitors. A new class of ceramic capacitors have been developed based on perovskite oxide dielectrics with excellent properties, *e.g.*: low dissipation loss, DC-field-insensitive, and temperature-independent high permittivity [1]. On the other hand, polymer-based composite films of polyvinyl alcohol and polyaniline modified carbon nanofibers have been produced for polymer composite capacitors with high dielectric constant and flexibility [2]. In this study, we utilize graphene oxide (GO) nanomaterial as an interesting dielectric material. The GO is one of the derivatives of graphene. Graphene is a nanomaterial that has excellent electrical conductivity. The derivatives of graphene, including reduced-GO (rGO), can be an alternative to help improve the performance of various energy storage devices [3]. One way to improve the dielectric property of the GO, which sets apart from the traditional capacitors, is via mild reduction possibly into rGO and at the same time retaining the insulator nature of the GO [4].

Graphite can function as an electrical conductor due to the delocalization of electrons between surfaces [5]. One layer of graphite is called graphene or in other words, graphite is an arrangement consisting of many layers of graphene where the layers are jointed together by the weak van der Waals forces. Graphene is obtained from exfoliating the previously reduced GO structure, which several very thin layers remain [6]. The two-dimensional (2D) structure of graphene covalent bonds has excellent optical, electrical, and mechanical properties. Therefore, graphene has the potential to be applied in terahertz transistors, ultrafast photodetectors, flexible touchscreens, sensors, and others [7]. GO material as a derivative of graphene can be obtained from graphite, prepared chemically by means of oxidation and exfoliation then undergoes oxidized modification from the basal plane [8]. Further reduction of oxygen content in the GO due to chemical, thermal, and other processes produces rGO [9].

GO can be produced via various methods. The most widely used method for synthesizing GO is the chemical oxidation of graphite, such as the Brodie method, Staudenmaier method, Hummer's method, and modified Hummer's method. There are advantages and disadvantages to each method used, but among the types of methods that have been mentioned, the modified Hummer's method is a fairly good method because it is commonly used and produces better GO compared to other methods.

In this article, GO samples are synthesized using the Hummer's method. Several characterizations are carried out

to show the properties of the samples that have been produced. The samples that have been characterized are then used to produce capacitors. In this case, the capacitors are made to compare the capacitance values using graphite and GO as the dielectric materials. The influence of the layer structure and thickness of the dielectric materials can also affect the capacitance value because during the production process, the coating and thickness of the GO can be changed. These changes can affect the electrical charge stored in the device.

2. Research method

This study aimed to produce GO via the Hummer's method. Then the GO sample obtained was characterized using the X-ray diffraction (XRD), ultraviolet-visible (UV-Vis) spectrophotometer, and Fourier transform infrared (FTIR) spectrometer. The GO was then used as a dielectric material in the parallel plate capacitors. Hence, the procedure in this study consisted of five steps, *i.e.*: i) preparing the GO sample from graphite powder; ii) characterizing the GO sample using XRD, UV-Vis, and FTIR; iii) producing the parallel plate capacitors with GO and graphite as the dielectric materials; and iv) testing the parallel plate capacitors produced using a multitester. The procedure of this study can be described as follows.

2.1. GO synthesis

One of the common methods to produce GO was the Hummer's method. This method was a chemical method that involved oxidation with sulfuric acid (H_2SO_4-Alfa Kimia; 98% purity), nitric acid (HNO_3-Alfa Kimia; 65% purity), and potassium permanganate ($KMnO_4$-Alfa Kimia; 99% purity). These reagents were used without further purification. The precursor material utilized was graphite (Sigma Aldrich; 99.9% purity) powder. In general, the GO was synthesized following the method given in Ref. [10]. The process of oxidation of graphite into graphite oxide was carried out by the Hummer's Method. The synthesis process was started by dissolving 1.0 g of graphite powder and 0.5 g of $NaNO_3$ with 25 ml of H_2SO_4. The solution was stirred for 1 hour. Next, 3 g of $KMnO_4$ was added slowly in an ice bath for 2 hours at a temperature below 20°C. After that, the solution was transferred into a heat bath and stirred for 20 hours at 40°C. The solution was then slowly added with 200 mL of H_2O. After 1 hour, 5 mL of H_2O_2 was added with continuous stirring for 1 hour. The final stage was adding 5 mL of HCl into the solution and stirred for 1 hour. After stirring, the beaker glass containing the solution was covered with plastic wrap and then left to settle. The solution that did not precipitate was discarded and H_2O was added. The mixture was put into 8 centrifuge tubes at 3080 rpm and rinsed every 15 minutes until the pH approached neutral. The precipitate was heated in the oven at 100°C for 38 minutes to remove the water content so that it becomes graphite oxide powder. The exfoliation of graphite oxide into GO was carried out by ultrasonication in an ultrasonic bath for 30 minutes. From this process, the GO dispersion solution was produced.

2.2. Manufacturing the capacitor using graphite material

The capacitors were made of aluminum plates with graphite material sandwiched between the plates. 2.0 g graphite powder mixed with 10 mL of H_2O. The graphite solution was stirred for 1 minute and then thinly coated on 2 aluminum plates. After that, activated carbon was sprinkled on both aluminum plates and allowed to dry for 30 minutes. One of the aluminum plates was given one layer of tissue and dripped with electrolyte solution with a mixture of 1.4 g of Na_2SO_4 and 10 mL of H_2O. The aluminum plates were then screwed and wired on each side. The capacitance value of the capacitors was measured using a multitester.

2.3. Manufacturing the capacitor using GO material

The capacitors were made of aluminum plates with mixture of GO and graphite materials sandwiched between the plates. Four aluminum plates were coated with GO material as much as 0.2 g and 0.8 g. The layers were allowed to dry for 30 minutes and then coated with graphite material. After drying and sprinkling with activated carbon, two aluminum plates (with different GO masses) were given tissue and dripped with the electrolyte solution. The aluminum plates were screwed and wired on each side, producing two parallel plate capacitors with GO masses of 0.2 g and 0.8 g. The capacitance value of the capacitors was measured using a multitester.

2.4. XRD characterization

XRD characterization was performed on the GO sample that has been synthesized. The purpose of the XRD characterization was to know the crystalline phase of the GO produced [11]. The XRD test to analyze the crystalline structure of the GO sample used Rigaku Miniflex 600 with X-Ray source CuKα (wavelength 0.15046 nm).

2.5. UV-Vis characterization

The optical properties of the GO sample can be studied through UV-Vis spectrophotometer characterization [12]. Measurement of the absorption spectrum of the GO samples used UV-Vis spectrophotometer (Ocean Optics USB4000) in the wavelength range of 200 nm to 800 nm. In this measurement, the GO sample was made in the form of a dispersed solution.

2.6. FTIR characterization

FTIR characterization was carried out to determine the functional groups in the GO sample by measuring the transmittance (%) or absorption of the GO sample. The FTIR test to

determine the functional groups in the GO sample used Shimadzu UV-3600 plus device.

3. Results and discussion

The XRD is a thin film characterization method used to determine the crystal structure and lattice parameters of materials. Moreover, tests using XRD aim to determine the crystallization phase of the GO material. The test is carried out with an angle range of 2θ between 4° to 80° with a wavelength of 1.54 Å.

The results of sample testing using XRD as shown in Fig. 1 show that two peaks are formed at 2θ of 12° and 43°. Based on previous research conducted by Yasin *et al*. [13] and Siburian *et al*. [14], the GO material has a characteristic peak (001) at 2θ of 12°. This peak corresponds to the d-spacing of the GO sheets of 0.75 nm [13,14]. Furthermore, a small bump (red circle in Fig. 1) appears at 2θ of 23° to 24°, which may indicate a transition from GO towards rGO. This may happen as the oxygen content in the GO sample is reduced, hence improving the electronic property of the samples, potentially increasing their dielectric performance [4, 15]. The reduction of the oxygen content in the GO sample is caused by the heating process of the GO inside the oven. However, based on Fig. 1, apart from the small diffraction peak indicating a transition from GO to rGO, there are also other peaks. This means that there are still impurities in the sample. Through the Hummer's method process, excessive graphite oxidation and incomplete reduction can cause defects and reduce the intrinsic properties of the GO [16]. As indicated by the diffraction peaks, impurities may appear in the sample. This impurity causes the resulting material to have lower conductivity, which in turn improves its dielectric property.

Next, UV-Vis spectrophotometry is a measurement of the interaction between electromagnetic radiation with atoms of a substance. The bound and unbound electrons are excited in a certain frequency region, which corresponds to that of ultraviolet and/or visible (UV-Vis) light. The UV-Vis absorption spectrum is about 200 nm to 800 nm. The UV spectrum

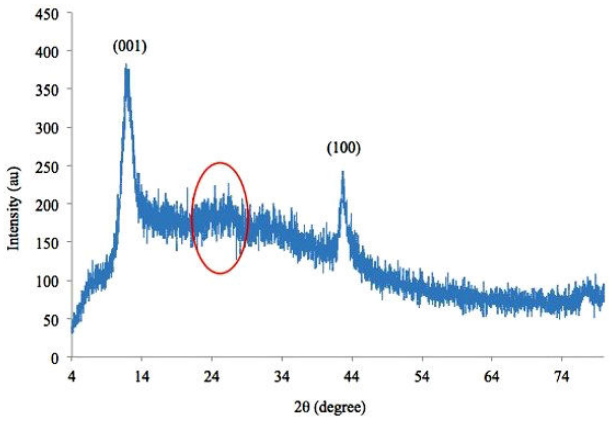

FIGURE 1. Diffraction pattern of the GO sample.

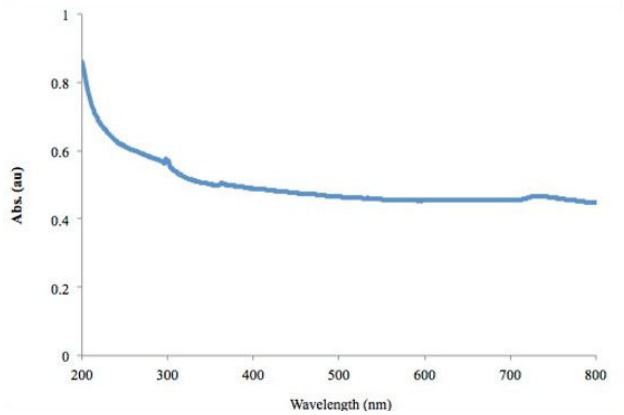

FIGURE 2. Absorption spectrum of the GO sample.

includes the UV region of 190 nm to 380 nm, while the visible (Vis) region is 380 nm to 780 nm. The measurement using the UV-Vis spectrophotometer is based on the relationship between the absorption and the wavelengths of the electromagnetic radiation. The UV-Vis characterization result of the GO sample may be observed in Fig. 2.

In general, the commonly accepted absorption spectrum of the GO is a peak occurring at 230 nm to 270 nm associated with the $\pi \rightarrow \pi*$ transition of the C=C bond [17]. In this case, it may be inspected in Fig. 2 that there is no peak present in the absorption spectrum of the GO sample at 230 nm to 270 nm. However, the absorption spectrum in Fig. 2 is in fact in accordance to the GO absorption spectrum in Ref. [18]. The absorption spectrum indicates the existence of ordered structure of GO, which may be due to the carbon rings in the basal planes of the GO. Moreover, a shouldering peak occurring at around 300 nm indicates electronic transition $n \rightarrow \pi*$ of the oxygen functional groups of the GO. From Fig. 2, the Tauc plot can be generated to determine the energy band gap (E_g) of the GO. This can be observed in Fig. 3. It is clearly seen from Fig. 3 that the E_g of the GO sample is about 3.9 eV,

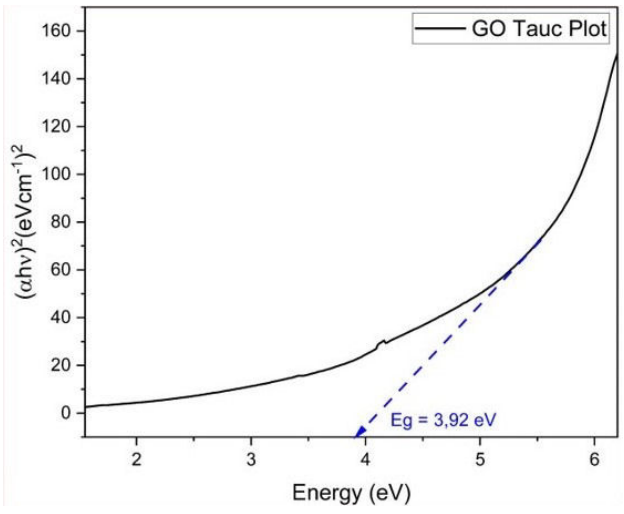

FIGURE 3. Tauc plot result of the GO sample.

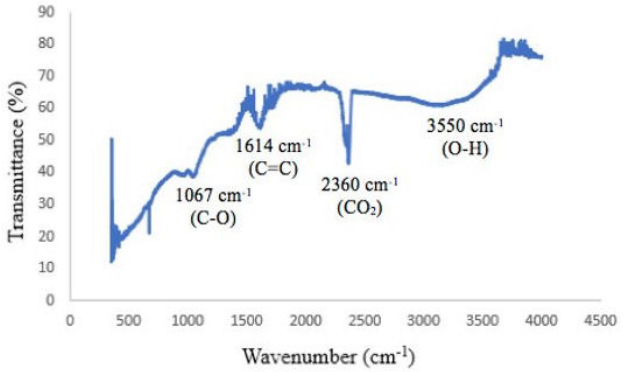

FIGURE 4. IR transmission spectrum of the GO sample.

which again is in accordance to the E_g in Ref. [18]. This shows that the GO sample obtained in this study is an insulator in nature and therefore can be used for dielectric material in capacitors.

Each molecule in a sample absorbs a certain amount of radiation energy. Radiation is absorbed through the sample, such that the molecules can absorb the radiation energy, which can be detected by the FTIR spectrometer. This FTIR result plots the amount of IR radiation transmitted through a sample as a function of the wavenumber of the radiation. This plot is called an IR spectrum, which gives important information about the functional groups of the sample. FTIR is used to identify functional groups of samples. The vibration of the molecules is caused by interactions in the form of energy absorption when IR electromagnetic radiation is applied to a material [19].

Analysis of the functional groups aims to determine the molecular bonds that exist in the GO sample. Figure 4 shows the transmittance spectrum of the sample and there are four functional groups detected. The GO sample produces peaks of C-O, O-H, C=C, and CO_2 bonds at wavenumbers of 1067 cm^{-1}, 3550 cm^{-1}, 1614 cm^{-1}, and 2360 cm^{-1}, respectively. The CO_2 functional group can come from the environment attached to the graphene surface. The results of the FTIR spectrum pattern show that the main molecular bonds owned by the GO sample are identified as C=C, C-O, and O-H bonds. The C=C functional groups indicate the formation of a hexagonal structure of carbon atoms arranged into GO layers. The appearance of the oxygen atoms is due to the oxidation process during the Hummer's method in the exfoliation process. Moreover, this confirms that the GO sample contains oxygen, and hence shows that GO has been successfully produced [20, 21].

The capacitor is made using thin aluminum plates measuring 8×5 cm^2. There are three variations of the capacitors made in this study, namely capacitors using graphite and GO with mass variation of 0.2 g and 0.8 g. The layers between the aluminum plates of the first capacitor used electrodes made from graphite, activated carbon, tissue, and electrolyte. Activated carbon is produced from heating GO, which functions to help the graphene layers increase its absorption for capacitance and electrical energy. As for the capacitors using GO, the layers are the same except for these capacitors the layers are given GO with masses of 0.2 g and 0.8 g, respectively. Both sides of the capacitor plates are attached to one cable, which is then tested using a multitester (see Fig. 5). This test was conducted to find out the capacitance of the capacitors.

The results of the capacitor testing can be shown in Fig. 6. The capacitor material produced in this study uses electrodes made from GO and activated carbon. The use of activated carbon aims to help the GO material to increase ion absorption for capacitance and electrical energy. The first capacitor uses graphite (GO with a mass of 0 g). The amount of capacitance produced is 0.000038 μF, which can be concluded that the ability of the capacitor to store charge is relatively weak. The second capacitor is using GO with a mass of 0.8 g. The value of the capacitance produced is 0.000041 μF. So it can be concluded that the ability of the capacitor to store charge

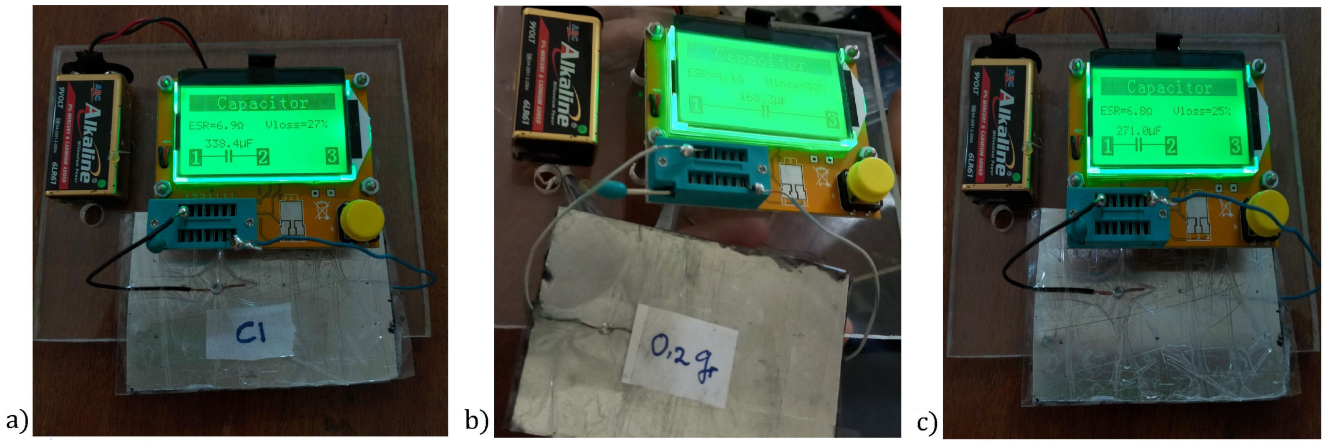

FIGURE 5. Measuring the capacitance of the canuapacitors with graphite a); GO with a mass of 0.2 g b); and GO with a mass of 0.8 g c).

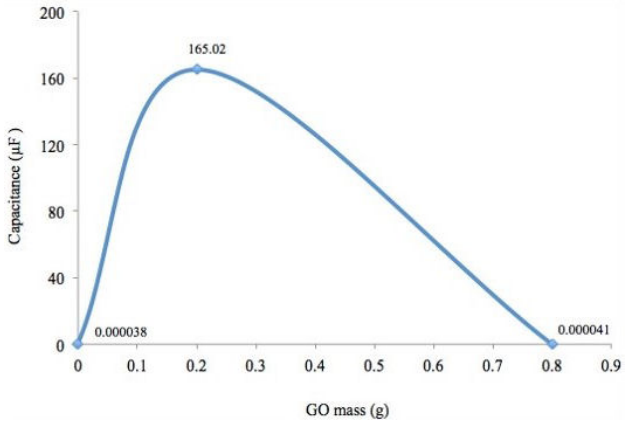

FIGURE 6. Test results of the capacitor capacitance.

is still relatively weak, but higher than the graphite capacitor. The third capacitor is using GO with a mass of 0.2 g. The value of the capacitance produced is 165.02 μF. Hence, the ability of the capacitor to store charge using GO with a mass of 0.2 g is much higher than the capacitors with graphite and GO with a mass of 0.8 g. The increase of the capacitance values for the capacitors containing GO is caused by the high E_g value of the GO sample as an insulator. Hence, the insulator nature of the GO sample increases the capacitance values of the capacitors. The insulating property of the GO sample can be attributed to the oxygen content in the sample, *i.e.*: C-O, O-H, and CO_2 as evidenced from the FTIR characterization result.

4. Conclusion

The GO material as a dielectric material for parallel plate capacitors has been produced. The synthesized GO has been characterized using XRD, UV-Vis, and FTIR. The result of the UV-Vis test shows a shouldering peak around 300 nm indicating the oxygen functional groups of the GO sample produced. Then the result of the XRD characterization indicates the occurrence of GO material with an intensity peak at 2θ of 12° and a transition from GO to rGO with a small bump at around 2θ of 23° to 24°. The FTIR characterization shows the functional groups of C-O, O-H, C=C, and CO_2 bonds at wavelengths of 1067 cm^{-1}, 3550 cm^{-1}, 1614 cm^{-1}, and 2360 cm^{-1}, respectively. The test results of the GO sample as the dielectric material for the parallel plate capacitors show that the ability of the capacitors to store charge is the highest for GO with mass of 0.2 g with a capacitance value of 165.02 μF. An interesting and necessary further study includes determining the thickness of the GO sample using morphological characterizations, *e.g.*: scanning electron microscope (SEM) and atomic force microscope (AFM).

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