Synthesis of Carbon Dielectric Composite from Candlenut Shell \textit{(Aleurites moluccana)}

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Abstract. Dielectric materials have attracted attention cause their potential applications such as energy storage devices and sensors. The biggest issues in the synthesis of dielectric materials reside in improving dielectric properties. In our work, a carbon dielectric composite material was successfully synthesized with PEG 4000 and PEG 6000 matrices respectively. The dielectric properties of candlenut shell carbon based composites filled with PEG 4000 and PEG 6000 were investigated. Conductivity of carbon decreases along with the addition of the matrices. The lowest conductivity value was obtained from Carbon / PEG 6000 material (0.52 – 0.91) x 10$^{-8}$ S / cm in the measurement range of 10$^3$ Hz - 10$^4$ Hz. This is due to the small carbon porosity (33.14%) and the addition of the matrices causing a decrease in the reactive surface area of the carbon.

1. Introduction
Capacitors are one of the electronic devices that can quickly respond to changes in direct current (DC) caused by variations in the intensity of sunlight on solar panels and have the expectation of a relatively longer usage time than batteries because they do not depend on reduction-oxidation reactions [1]. Besides that photocapacitor also has high quantum conversion efficiency with output voltage reaching 0.45 V for open circuit voltage which is still in the range of output voltage of conventional solar cells (battery-based solar cells) with a capacitance value of 0.69 F.cm$^{-2}$ [1]. Photocapacitor modification using three electrodes consisting of TiO$_2$ mesoporous electrodes, carbon-coated and electrolyte solutions obtained output voltage 0.8 V or five times greater than the photocapacitor of the two previous electrodes [2,3].

The prototype hybrid energy converter fabrication and integrated energy storage devices using ZnO nanowires (NWs) and graphite as electrodes show that the capacitance of the supercapacitor fiber comes from ion absorption effectively on the electrolyte / ZnO NWs interface. In addition, the unique structure of ZnO NWs and graphite-based fibers as active materials and electrodes is very useful and can be used flexibly [4]. A new generation of solar energy conversion devices such as DSSCs, PSCs and PVSCs with planar and fiber shapes with overall superior properties has efficiency that exceeds 10% of supercapacitor efficiency [5].

Development of Matrix Composites Polymer based capacitor electrodes began to be developed because of their superiority and high applicable value. Synthesis of carbon nanopores from coconut shell as electrodes on supercapacitors with activator of polyethylene glycol (PEG), polyvinyl alcohol (PVA) and silica nanoparticles (SiO$_2$) produce different pore distributions. PEG and PVA activators
show a fairly even pore distribution on the carbon surface while silica activators have not been effective enough to form pores [6]. Synthesis of capacitor electrodes from candlenut shell (Aleurites moluccana) using polyaniline (PAni) activator with carbon activation temperature variations obtained the highest conductivity of $5.7 \times 10^{-3}$ Sm$^{-1}$ with a capacitance value of 10.52 μF [7]. Analysis of the effect of the zinc (Zn) reducing agent on the capacitive properties of the graphite material supercapacitors produces the best results in addition of 0.8 grams of Zn with a conductivity value of 2,906 S.cm$^{-1}$ and a capacitance value of 90.623 F / gram with thin sheet morphology [8]. The increase in mass fraction on TiO$_2$ in TiO$_2$/C composites resulted in an increase in the supercapacitive capacitance value. The role of TiO$_2$ in increasing the dielectricity of supercapacitors is due to the conductive nature of carbon due to the activation processes.

2. Experimental Procedures

2.1 Candlenut Activated Carbon Synthesis
Synthesis begins with preparation of the candlenut shell (Aleurites moluccana) which includes two stages, namely the cleaning process and the drying process. In the cleaning process, the candlenut shell is soaked in water so that the remaining impurities such as soil and fruit flesh can still be dissolved. To clean the remaining flesh of the seeds that are still attached in addition to using water, a brush is also used so that the remaining meat from the seeds is quickly peeled off. After the candlenut is washed clean, the drying stage is carried out using sunlight. The drying process depends on the intensity of the sun every day. Dry candlenut shells are then carbonated using a furnace at a temperature of 500°C for 30 minutes. The candlenut shell charcoal from the carbonization process is then crushed using a mortar then filtered using a 200 mesh filter. Carbon powder measuring 200 mesh is then soaked with phosphoric acid for one day. The carbon immersion results are separated by the filtrate and then activated by heating with the furnace at 650°C for 2 hours. The activated carbon is washed using distilled water to reach a neutral pH then dried using an oven at 100°C for 1 hour.

2.2 Synthesis of Carbon Dielectric Composite
Synthesis carbon dielectric composite based candlenut shells started by mixed candlenut activated carbon (AC) with polymer matrix polyethylene glycol 4000 (P4) as (ACP4) and polyethylene glycol 6000 (P6) as (ACP6) with a mixed ratio of 0.2 grams of activated carbon: 0.4 grams of polymer matrix respectively. The mixture compacted using a hydraulic press machine to form carbon dielectric composite tablets and sintered at 40°C with a hot plate for 30 minutes.

3. Results and Discussion

3.1. Candlenut Activated Carbon Microstructure
The activated carbon from the candlenut shell has been successfully synthesized. From the microstructure testing data using SEM obtained the surface morphology of the candlenut shell activated carbon shown in Figure 1. This micrograph has a magnification of 3000 times and a pixel scale conversion factor of 0.05 μm / pixel. The activated carbon pore has a darker color than the color of the matrix and the pore geometry is close to the circle. By using ImageJ software analysis of pore characteristics is obtained the small carbon porosity about 33.14% [9].
Figure 1. Micrograph of the candlenut shell activated carbon

3.2. Diffraction data Analysis
Analysis of diffraction data on activated carbon and activated carbon precursors is shown in Figure 2. The meaning of activated carbon precursors is candlenut shell charcoal after experiencing immersion with phosphoric acid (chemical activation). The main components of the candlenut shell are lignin, cellulose and hemicellulose. In general, lignin and hemicellulose have an amorphous structure, whereas cellulose in part has a crystalline structure. The presence of cellulose in the candlenut shell allows it to have a crystalline structure. Whereas in the charcoal the crystalline structure is formed due to the hexagonal layer of carbon formed during the carbonization and thermal activation processes.

Figure 2. Diffraction data of activated carbon (raw) precursors and activated carbon from candlenut

From the precursor diffraction data and activated carbon generally an amorphous structure was formed in both samples. This is due to the ability of phosphoric acid to break down cellulose in the candlenut shell charcoal. It can be observed that what has been characterized by the X’Score HighScore Plus and Match software there is a crystalline phase at a value of 2θ which is thought to be the hexagonal phase of graphite (*) and cubic diphosphate silicon (#). The presence of graphite hexagonal phase is possible to form during the carbonization process. The presence of the silicon diphosphate cubic crystalline phase indicates that during the chemical activation process phosphate compounds react with silicon to form a cubic crystal structure. However, after thermal activation at a temperature of 650°C, this structure disappears presumably at this temperature the bond between silicon and phosphate becomes unstable and then breaks back into silica and phosphate derivative solids in the form of ash.
3.3. Electrical Properties of Carbon Dielectric Composite

The electrical conductivity value in the sample is obtained from the results of resistance measurements using frequency variations ranging from $10^3$ Hz to $10^4$ Hz. This aims to see the electrical conductivity properties for various types of frequencies used. In general, the increasing frequency value that is applied, the value of the conductivity of the sample will also increase as shown in Figure 3. This increase in frequency causes the number of charge carriers in the material to increase, resulting in a high transfer of charge to the interface area resulting in increased material conductivity.

![Conductivity Graph](image)

**Figure 3.** Graph the conductivity value for frequency changes

Table 1 shows the range of electrical properties which include the values of resistance, resistivity and conductivity of samples in the frequency range of $10^3$ Hz - $10^4$ Hz. It is seen that the five samples have conductivity values in the order of $10^{-8}$ S/cm. Based on the conductivity value the material is divided into three types, namely insulating material ($10^{-18}$ - $10^{-8}$ S/cm), semiconductor material ($10^{-8}$ - $10^{-3}$ S/cm) and conductor material ($10^{-3}$ - $10^8$ S/cm) [9]. This shows that the samples that were successfully synthesized have transition properties between semiconductor material and insulators. It is clear that the material successfully synthesized is not suitable if used as an electrode because of its low conductivity value. This means that this material cannot be used as a charge storage device but on the contrary this material can be used as a semiconductor material or an insulating material.

| Sample | Resistance ($10^7$ Ω) | Resistivity ($10^7$ Ω cm) | Conductivity ($10^{-8}$ S/cm) |
|--------|------------------------|---------------------------|------------------------------|
| AC     | 3.18 – 4.01            | 8.89 – 11.20              | 0.89 – 1.13                  |
| P4     | 1.57 – 3.04            | 4.81 – 9.30               | 1.07 – 2.08                  |
| P6     | 0.61 – 0.73            | 2.03 – 2.35               | 4.26 – 4.91                  |
| ACP4   | 1.40 – 1.91            | 8.85 – 12.04              | 0.83 – 1.13                  |
| ACP6   | 1.90 – 3.31            | 11.03 – 19.20             | 0.52 – 0.91                  |

The conductivity value of candlenut activated carbon is obtained at ($0.89 - 1.13$) $10^{-8}$ S/cm. In general, measurements of polymer conductivity that are experiencing exposure using candlenut shell activated carbon depend on the concentration of activated carbon (dopant), homogeneity of dopants in polymers and polymer morphology and dopants, and chemical reactions that occur between dopants and...
polymers [10]. The conductivity value of PEG 6000 is higher than the conductivity value of PEG 4000. However, after experiencing a drop in the conductivity value the Carbon Dielectric Composite's sample decreases. This is presumably due to the distribution and composition of activated carbon added. The distribution of this distribution can be caused by the pore diameter and porosity of the activated carbon. Activated carbon that has been synthesized has porosity of 28.76% - 33.14% which is still relatively low. The higher porosity value of an activated carbon allows an increase in surface area so that more and more diffusion of the polymer on the pore surface of the activated carbon.

4. Conclusion
The candlenut activated carbon synthesized using phosphoric acid activator and activated thermally at 650°C has a fairly evenly distributed pore with an average pore diameter of 3 μm with a percentage of porosity ranging from 28.76% to 33.14%. The electrical properties of composite based on candlenut shell activated carbon used PEG 4000 and PEG 6000 as matrix have conductivity values in the testing range of frequencies of $10^3$ Hz to $10^4$ Hz each ranging from $(0.83 - 1.13) \times 10^{-8}$ S/cm and $(0.52 - 0.91) \times 10^{-8}$ S/cm is dielectric materials and suitable as carbon dielectric composite candidate.

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