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The Effect of rGO Mass Composition on The Performance of Activated Carbon/rGO Supercapacitor Electrode Based on Coconut Shell (*Cocos nucifera*)

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Abstract. Activated carbon (AC) is composed of reduced graphene oxide (rGO) and has been synthesized from natural material that is coconut shell (*Cocos nucifera*) by dry mixing method. The purpose of this research was to know the effect of adding rGO mass on the electrode to the specific capacitance value. The rGO mass variation used was 10-25 wt%. AC/rGO electrodes were tested by cyclic voltammetry to determine their specific capacitance values at a scan rate of 100 mV/s with Sodium Sulphate (Na_2SO_4) 1 M in the electrolyte solution and a potential range at 0-1 Volt. The electrode by 90 wt% AC composition and 10 wt% rGO had a higher specific capacitance value of 6.26 Fg^{-1} than other electrode mass compositions, as well as the activated carbon individual electrodes having a specific capacitance value of 3.38 Fg^{-1}. Therefore, it could be concluded that the specific capacitance of the electrodes could be increased by the addition of rGO material.

1. Introduction

Supercapacitor is an energy storage system which is capable to provide high power storage in a short time [5,9,19] and has a cell capacitance range of 0.043 F - 2,700 F [13]. Generally, a supercapacitor consists of a pair of electrodes, a separator and an electrolyte solution [1]. Determining the electrical properties of the supercapacitor requires selection of the right material as a supercapacitor electrode. Electrodes are electrical conductors that have the ability to absorb and release electrons from their surfaces. The storage process occurring on the electrode surface will affect the specific capacitance value [15]. Materials that have already begun to be used as supercapacitor electrode materials include activated carbon, *graphene* and reduced Graphene Oxide [3,7,19]. Reduced Graphene Oxide (rGO) is a *graphene* oxide material in which the carbon atoms of *graphene* undergo oxidation and reduction processes. In the process of oxidation, there are several oxygen and hydrogen atoms bonded to carbon atoms called graphene oxide. Meanwhile, in the process of reduction, the release of several bonds of graphene oxide occurs so that it obtaines structure that almost resembles *graphene*. The result of this reduction process is called reduced graphene oxide (rGO). The difference between *graphene*, *graphene oxide* and rGO is the content of the O. *Graphene* atomic group is only composed of carbon atoms while *graphene oxide* and rGO have an atomic O group. The O group on *graphene oxide* is more (41-50%) than rGO (13-22 %) [14]. The synthesis of *graphene* and activated carbon composites as supercapacitor electrodes is performed by ultrasonification, hydrothermal carbonation and chemical activation resulted in a specific capacitance of 210 Fg^{-1}. The capacitance of the *graphene* composite and the activated carbon is greater

than the capacitance of the pure activated carbon electrode of 76 Fg^{-1}. It suggests that with the addition of graphene can improve the process of storing the charge of the electrode [19]. In subsequent studies, the activated carbon/reduced Graphene Oxide (rGO) composites were used as electrodes through hydrazine method [7]. The synthesis of the composite electrode was performed by using different variations of rGO mass composition i.e. 10 - 50 wt% rGO which, in the mass of 20 wt% rGO, obtained the best capacitance value of 181 Fg^{-1} compared to the mass of the other electrodes. The activated carbon/ rGO composite electrode also has a higher capacitance value than the pure activated carbon electrode [7]. A research [6] concerning the manufacture of rGO from coconut shell was then compiled with ZnO as a supercapacitor electrode, in which the composite mass variation was rGO-ZnO (1: 1, 1: 2 and 2: 1). The result of the research obtained the best capacitance value on rGO-ZnO composite with the ratio of 1: 2 composition of 0.613906 Fg^{-1}, where the process of voltametric characterization at scan rate 100 mVs^{-1} by using electrolyte solution KOH 1 M [6]. Based on the aforementioned researches, the material used was relatively expensive [7,19], so this research proposed an innovation by using simpler and easier method in making activated carbon /rGO (reduced Graphene Oxide) composite without the need of expensive solvent. The method of making electrode in this research used dry mixing method. Dry mixing method is a method of mixing the material in the form of a powder with a certain length of time without using solvent and it is done in the outside air and used when the material does not easily experience oxidation in the outside air [8].

2. Materials and Methods

2.1. Materials

The material used was from the natural coconut shell (*Cocos nucifera*). All chemicals used were pure analys and used without purification, including Sodium Oxide (NaOH 0.5M) as chemical activator, Chloride Acid (HCl 1M), Sodium Sulphate (Na_2SO_4 1M) as electrolyte solution, Poly Ethylene Glycol (PEG) 4000 as a binder and deionize water.

2.2. Synthesis of Activated Carbon

The activated carbon is synthesized from coconut shell using dehydration method, carbonation and chemical activation. The coconut shell was dehydrated in ± 7 days then carbonized at 400 ^{0}C and smoothed. Afterwards sieved on 200 mesh sieve then activated using 0,5 M NaOH activator and soaked for 24 hours with carbon mass ratio: NaOH of 1: 3. Subsequently, the carbon bath was calcined at 800 ^{0}C for 5 hours, after which it was washed with 0,5 M HCl and deionized water to a pH of 7 [12]. The activated carbon powder is then dried at 110 ^{0}C for 1 hour to remove the water content in the powder.

2.3. Synthesis of reduced Graphene Oxide (rGO)

Reduced Graphene Oxide (rGO) is synthesized from coconut shell with dehydration, carbonation and calcination. The coconut shell was dehydrated for ± 7 days then carbonized at 400 ^{0}C. After it was calcined with a temperature of 400 ^{0}C for 5 hours then it crushed into powder. Then, the rGO powder was washed with deionize water until the powdered sludge was obtained [10]. To remove the water content, it was then dried with a temperature of 110^{0}C for 1 hour.

2.4. Synthesis of Supercapacitor Electrode

The supercapacitor electrode was made by dry mixing method on two main ingredients, activated carbon and reduced Graphene Oxide (rGO). The activated carbon/rGO composite electrode was composed of several mass variations i.e. 10 wt% to 25 wt% rGO. Each of the activated carbon powder and rGO was mixed simultaneously with Poly Ethylene Glicol (PEG) 4000 as a powder binder at a ratio of 1: 1 [18]. The mixture was crushed by using alu mortar for 1 hour to obtain a homogeneous powder mixture. Furthermore, the electrode powders were compressed using a pressure of 1.5×10^6 Pa to form a pellet-like electrode. Compaction method is a process of powder compaction into a sample with a certain shape according to the mold [8].

3. Instruments

The activated carbon powder and reduced Graphene Oxide (rGO) were characterized using X-Ray Diffraction with an angle of $2\theta = 10^0$-50^0 of 0,5-1,0 grams to understand the phase and material structure. XRD test result was presented in graphic characteristic of XRD. The activated carbon /rGO electrode was characterized by Cyclic Voltemetry to know the electrochemical characteristics of the specific capacitance from the electrode. Cyclic Voltemetry (CV) was tested on all variations of composition of activated carbon/rGO mass with electrolyte Sodium Sulphate (Na_2SO_4 1M) solution, scan rate 100 mVs^{-1} and potential range at 0 - 1 Volt resulting voltamogram curve. The analysis of the electrode conductivity properties used Electrochemical Impedance Spectroscophy (EIS) method in the frequency range 100 kHz - 100 mHz with the result of Nyquist plot.

4. Result and Discussions

The results of the characterization of X-Ray Diffraction on the activated carbon and reduced Graphene Oxide (rGO) synthesized from the coconut shell (*Cocos nucifera*) is shown in Figure 1. The test used powder in which each sample was as much as 0,5 - 1,0 gram by using angle (2θ) in the range 10^0 - 50^0.

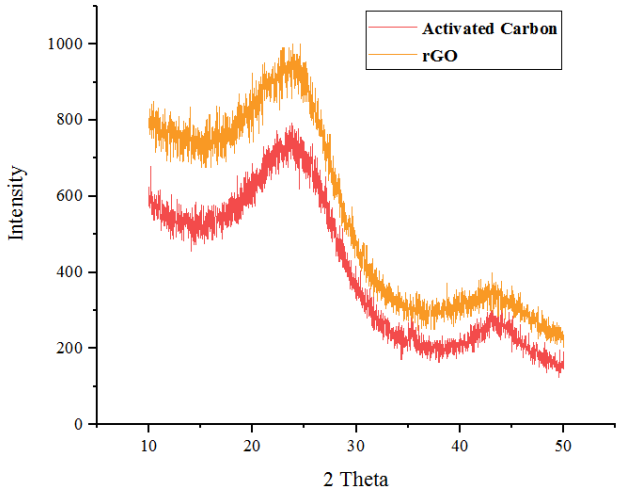

Figure 1. XRD patterns of activated carbon and reduced Graphene Oxide (rGO).

The XRD patterns in Figure 1 is the characteristic of activated carbon and rGO. The two wide diffraction peaks were at $2\theta = 20$ - 30^0 and $2\theta = 40$ - 50^0 indicating that the phase was amorphous, composed by carbon bonds irregularly so it could be used as adsorbent material [17]. The activated carbon from the coconut shell in this study showed that the diffraction peaks were at $2\theta = 35^0$, 43^0 and 45^0. While the pattern on the reduced Graphene Oxide (rGO) shows the diffraction peak at $2\theta = 24^0$ and 43^0 where there was the same peak characterization at $2\theta = 24^0$ and 44^0 [4]. The rGO phase formed was semi crystalline which was proved to have a higher intensity compared to activated carbon. The XRD intensity value was influenced by the crystallinity rate of a material, if the material was increasingly crystalline, the intensity got higher [17]. The results of the characterization of both samples showed the existence of a stacked peak and wide, whereas the difference was at the diffraction peak ie the 2θ angle formed and the intensity level.

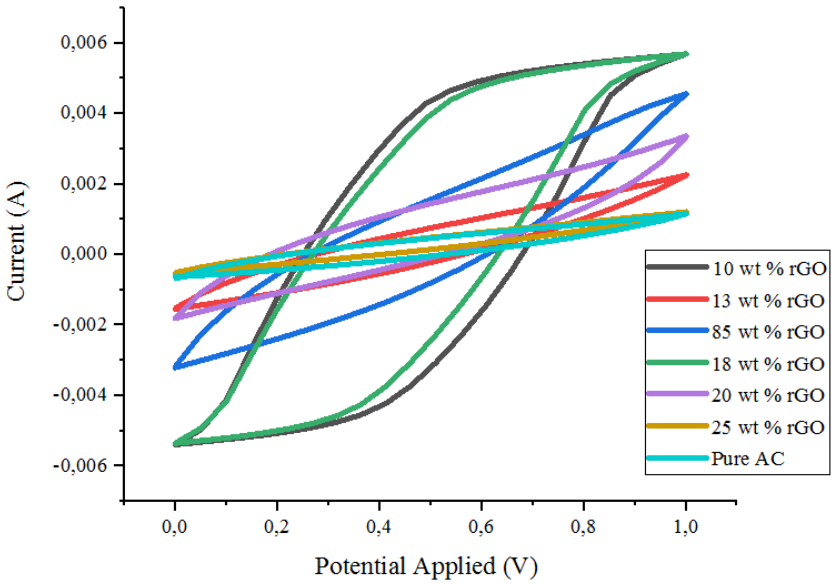

Figure 2. CV curve of pure AC electrode and composite electrode of activated carbon/rGO at different mass composition using scan rate 100 mVs^{-1} with potential range 0-1 V in 1M Na_2SO_4 electrolyte solution.

Cyclic voltametry was the most commonly used characterization test for determining the capacitance performance of the electrodes and the quantitative value of the electrode specific capacitances [5,19]. The CV curve of the activated carbon/rGO electrode was shown in Figure 2 at all variations of the mass composition using a scan rate of 100 mVs^{-1} and a potential range of 0 - 1 V in a 1M Na_2SO_4 electrolyte solution. The CV curve obtained had a different shape and area due to the capacitance value held on each of the different electrodes. The specific capacitance values of electrodes 10 wt%, 13 wt%, 15 wt%, 18 wt%, 20 wt% and 25 wt% rGO were obtained in the order of 6.26 Fg^{-1}, 5.27 Fg^{-1}, 3.87 Fg^{-1}, 4.24 Fg^{-1}, 3.71 Fg^{-1} and 3.88 Fg^{-1}. Meanwhile, the pure electrode capacitance value of pure activated carbon was smaller than all composite electrodes with 3.37 Fg^{-1}. The electrode capacitance value could be increased by the addition of rGO material [7], then the rGO material from the coconut shell could be used as composite material on activated carbon. The network structure was made by the incorporation of rGO material into the activated carbon electrode. It bridged the pores which were connected with the activated carbon particles conductively. Thus, the rapid electrolyte ion transport process was facilitated in the electrode [7]. Furthermore, it was found that the specific capacitance value decrease was caused by the presence of increasingly rGO composition. The decreasing capacitance value was caused by the composition of excessive rGO material. Most of the surface of the activated carbon was covered by rGO, which the ion diffusion process would be made in the pores of the electrode obstructed and the obtained low capacitance value would be affected [7]. If the pores of the electrodes were closed to the addition of too much composite material, then the electron absorbtion process would be inhibited. The magnitude of the capacitance value of the electrode was affected by that [3]. Therefore, the best electrode mass composition with the highest capacitance value was 90 wt% activated carbon composition and 10 wt% rGO.

The activated carbon/rGOelectrode 90-10 with the best capacitance value was tested by EIS or Electrochemical Impedance Spectroscophy in order to determine the conductive behavior of the electrode meterial [19].

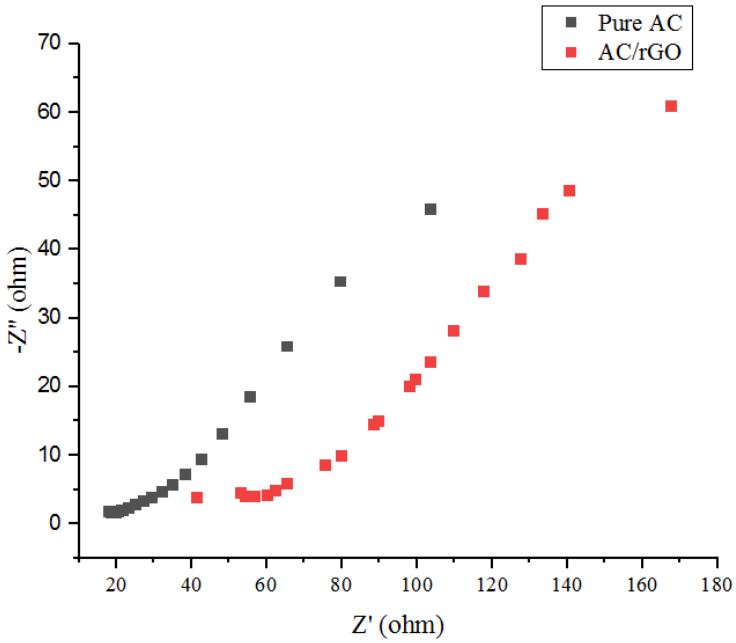

Figure 3. Nyquist plot of pure activated carbon and activated carbon / rGO electrode with a frequency range of 100 kHz - 100 mHz.

The Nyquist plot of the activated carbon electrode and the activated carbon/rGO electrode was shown in Figure 3. Figure 3 is an Electrochemical Impedance Spestroshopy (EIS) analysis with a frequency range of 100 kHz - 100 mHz. Nyquist plot consisted of component Z ' is a real impedance and Z " is an imaginary impedance. The Nyquist EIS plot in the drawing also consisted of 3 parts, the semicircle, the almost vertical line and the vertical line. Therefore, through the plot Nyquist, the obtained electronic resistance value (R_e), which was the lowest point and the charge transfer resistance (R_{ct}), were the difference of the highest value with the lowest value [2]. In Fig. 3 (a) the Nyquist plot was obtained from the activated carbon/ rGO composite electrode with R_{ct} of 13.0193 Ω and R_{tot} of 54.3093 Ω. Meanwhile, the pure activated carbon electrode obtained R_{ct} value of 2 Ω and R_{tot} of 18.9614 Ω. R_{tot} which were the total of R_e and R_{ct} [2]. In addition, Figure 3 shows that the diameter of the semicircle plot on the composite electrode was greater than that of the pure activated carbon electrode. When the diameter of the semicircle increased, the load transfer resistance (R_{ct}) [11] became higher. Therefore, the value of the quantitative charge transfer resistance in the activated carbon/rGO electrode was higher than the one of the pure activated carbon electrodes.

5. Conclusion

The best specific capacitance value was obtained at composite electrode with 90 wt% activated carbon composition and 10 wt% rGO ie 6.26 Fg^{-1}, while the specific value of pure active carbon electrolactivity was smaller at 3.38 Fg^{-1}. Therefore, the specific capacitance value on the activated carbon electrode could be increased with the addition of rGO mass.

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