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Specific capacitance of composite electrode activated carbon/rGO of coconut shell (*Cocos nucifera*) as supercapacitor electrode

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Abstract. The Activated Carbon/rGO supercapacitor electrode from coconut shell (*Cocos nucifera*) in this research has been successfully synthesized using dry mixing method. In this study the mass composition of composite used 10 wt% rGO and 90 wt% Activated Carbon. To find out the capacitance of the electrode was performed cyclic voltammetric characterization at a scan rate of 1-5 mVs⁻¹ in a 1 M Na₂SO₄ electrolyte solution. Electrode characteristic test used is BET and CV test. The CV characterization results show the best capacitance value occurs at scan rate of 1 mV/s ie 336 Fg⁻¹. At a low scan rate on cyclic voltammetry, the electrons have a long time to diffuse evenly to the electrode surface and the greater the charge transfer resulting in higher capacitance values. Conversely, the higher the scan rate, the electrons can diffuse rapidly and the charge transfer process on the electrode surface is very limited.

1. Introduction

The development of research on natural materials that have the potential as renewable technologies is increasing rapidly. One application of technology that is often used in life is the supercapacitor electrode. Many studies have succeeded in synthesizing supercapacitor electrodes with good electrochemical performance [1,2]. The supercapacitor is a device with an energy storage system that is able to provide high power storage in a short time [2,3,4] and has a cell capacitance range between 0.043-2.700 F [5]. In general the supercapacitor consists of a pair of electrodes, separator and electrolyte solution [6] which will make the process of ion transfer occur between the two electrodes. The process takes place where the voltage is applied to the supercapacitor, the ions in the electrolyte solution diffuse into the pores of the electrode. Charging ion that occurs between the surface of the electrode and electrolyte, forms two charged layers (double layers) with a very small separation distance [6]. In determining the electrical properties of the supercapacitor, it is necessary to select the right material as the supercapacitor electrode. Supercapacitor electrodes are electrical conductors that have the ability to absorb and release electrons from their surface. The storage process occurring on the electrode surface will affect the specific capacitance value [7]. The electrical properties of the supercapacitor are determined by the selection of appropriate materials. Electrodes are electrical conductors that are able to absorb and release ions from their surfaces. On the surface of the electrode, the accessibility of ions is influenced by the pore diameter, or indirectly in the form of pore volume. The smaller the pore size, the ion accessibility will be faster and will increase the efficiency of the surface area on the electrode. Electrodes that have a very large surface area will maximize the absorption of electrons, so with a very large surface area it



is possible to have a large capacitance value as well. The requirements of the supercapacitor electrode are to have low resistance, good electrolyte accessibility, good chemical and mechanical stability, and high capacitance values [8]. Supercapacitor electrodes can be made by using Activated Carbon materials [1,2]. One study that has synthesized reduced Graphene Oxide/Activated Carbon (GAC) composite electrodes with a specific electrode capacitance value was obtained using a cyclic voltammetry test (CV) at a scan rate of 1 mVs^{-1} with 1 M NaCl solution. Specific capacitance value of GAC composite electrode obtained the best capacitance value that is equal to 181 Fg^{-1} in mass composition GAC-20 (rGO-20 and AC-80) [1]. Furthermore, other studies conducted graphene and activated composite synthesis as a super-capacitor electrode with a simple method that is by ultrasonification, hydrothermal carbonation and chemical activation with KOH. Specific capacitance values obtained on Graphene/Activated Carbon electrodes of 210 Fg^{-1} at a scan rate of 1 mVs^{-1} , are higher than the Activated Carbon electrodes of 76 Fg^{-1} at a scan rate of 1 mVs^{-1} [2].

Based on several studies above the material used is relatively expensive [1,2] because it is not derived from natural materials. This research innovated using natural materials and simpler methods and easier in the process of making Activated Carbon/rGO (reduced Graphene Oxide) composites. This research was carried out without the need for expensive solvents. The method of making composite electrode is 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 the use of solvent and done in the outside air, used when the material is not easy to experience oxidation in the outside air [9]. So this research is able to make composite electrodes with affordable materials and simpler methods but are able to have good specific capacitance values as electrodes. Furthermore, in another study about Activated Carbon/PVDF composite electrodes using a simple mixing method with a composition of 10 wt% PVDF. Activated Carbon/PVDF composite electrodes are given a separator made of polymer electrolyte (PVA/ H_3PO_4) by a sol gel method and then characterized using cyclic voltammetry (CV) with scan rate variations at 5 mVs^{-1} , 10 mVs^{-1} , 20 mVs^{-1} , 25 mVs^{-1} and 50 mVs^{-1} . The results show the largest specific capacitance value of 86 Fg^{-1} at the lowest scan rate in 5 mVs^{-1} [10]. This research of Activated Carbon/rGO composite electrodes which had been synthesized was tested by BET (*Brunauer-Emmet-Teller*) to find out the pore size and surface area of electrodes and Cyclic Voltammetry (CV) using a scan rate variation of $1\text{-}5 \text{ mVs}^{-1}$ which refers to previous studies.

2. Materials and Method

2.1. Materials

The material used in this study is derived from natural ingredients, namely coconut shell (*Cocos nucifera*). Additional and supporting materials used in the synthesis of Activated Carbon/rGO composite electrodes are chemicals that are pure analytically and used without purification, including Sodium Oxide ($0,5 \text{ M NaOH}$) as a chemical activator, Chloride Acid (1 M HCl), Sodium Sulfate ($1 \text{ M Na}_2\text{SO}_4$) as an electrolyte solution, Poly Ethylene Glycol (PEG) 4000 as a binder and deionized water.

2.2. Synthesis of Activated Carbon

Method of making Activated Carbon from coconut shell using dehydration method, carbonation and chemical activation. The coconut shell is dehydrated under the sun ± 7 days, then carbonized at 400°C and smoothed. After that it was sieved on 200 mesh sieve then activated using $0,5 \text{ M NaOH}$ activator and soaked for 24 hours with carbon mass ratio:NaOH of 1:3. Subsequently, the carbon bath was calcined at 800°C for 5 hours, after which it was washed with 0.5 M HCl and deionized water until the pH reached 7 [11]. The Activated Carbon powder is then dried at 110°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 natural coconut shell (*Cocos nucifera*) using dehydration, carbonation and calcination methods. Coconut shells that have been prepared are dehydrated for ± 7 days then carbonated at 400°C . After that it is calcined with a temperature of 400°C for 5 hours and then mashed to become a powder. Furthermore, the rGO powder was washed with

deionize water to obtain a precipitate of rGO powder [12]. To remove the water content then dried with a temperature of 110 °C for 1 hour.

2.4. Synthesis of Supercapacitor Electrode

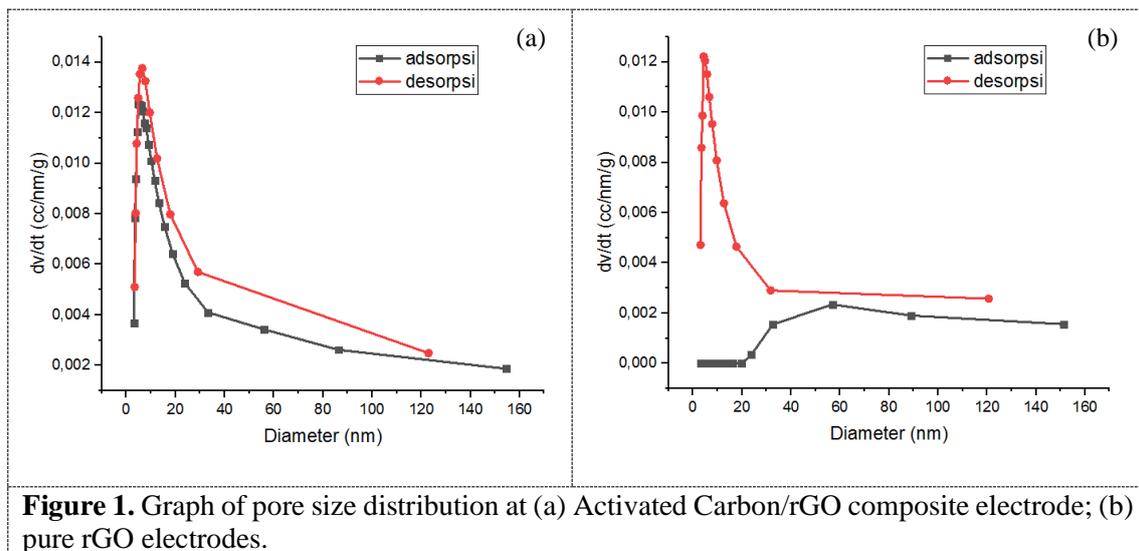
The Activated Carbon/rGO composite electrode as a supercapacitor was prepared by using dry mixing method on two main ingredients ie Activated Carbon and reduced Graphene Oxide (rGO), synthesized from natural material ie coconut shell (*Cocos nucifera*). The composition of the Activated Carbon/rGO composite electrode was used for 90 wt% Activated Carbon and 10 wt% rGO. Pre-synthesized Activated Carbon and rGO powders were mixed simultaneously with Poly Ethylene Glicol (PEG) 4000 as a powder binder at a ratio of 1: 1 [13]. The powder mixture was crushed by using alu mortar for 1 hour until a mixture of homogeneous powder was obtained without any clumps. Furthermore, the electrode powder is compressed using a pressure of 1.5×10^6 Pa to form a pellet-like electrode. The compaction method is a process of compacting powder into a sample with a certain shape according to the mold [9].

3. Instrumentation

The Activated Carbon/rGO electrode characterized by BET (*Brunauer-Emmet-Teller*) is a characterization commonly used to find out the pore size and surface area of the sample [11]. The characterization tools used are the NovaWin BET Quantachrome 11.0 engine contained in the Integrated Science Laboratory of the Faculty of Mathematics and Natural Sciences, Universitas Negeri Surabaya. Electrode samples in the form of 0.14 gram of powder were heated using temperature 300°C with nitrogen gas (N₂) sample for 3 hours. Subsequently the Activated Carbon/rGO composite electrode was characterized by Cyclic Voltmetry to determine its electrochemical characteristics. Cyclic voltametry (CV) is an effective method of determining electrochemical performance of the electrode material [1,2]. Most studies have used cyclic voltammetry tests as a characterization tool suitable for characterizing electrode capacitive behavior and can measure the specific capacitance values of electrodes [1,2]. So in this study electrode tested voltemetri Cyclic (CV) to know the value of its specific capacitance. The CV test used a scan rate variation of 1-5 mVs⁻¹ in 1M sodium sulfate (Na₂SO₄) electrolyte solution and a potential range at 0 - 1 Volt resulting in a voltammogram curve.

4. Result and Discussions

The Activated Carbon/rGO composite electrode has been synthesized from the coconut shell characterized using BET and CV. BET or *Brunauer-Emmet-Teller* is used to determine the physical characteristics of electrodes. The characterization used to determine the pore size and surface area of a material sample is the characterization of BET (*Brunauer-Emmet-Teller*). Pore size in an electrode can affect its electrochemical performance. Electrodes having large surface area will maximize the ion absorption process, there by allowing an electrode to have a large capacitance value [8]. In the BET testing process the sample electrode is heated and fed by nitrogen gas where the gas portion entering into the pores produces the adsorption curve, otherwise the gas coming out of the pores yields the desorption curve. The process is analyzed pore size and pore surface area using BJH or (*Barret-Joyner-Hallenda*) method. Based on the BET characteristic results, the pore size distribution curve is shown in Figure 1.



Based on the result of BET characteristic, the pore size distribution values of each sample are shown in Table 1.

Table 1. Pore size distribution ratio of Activated Carbon/rGO composite electrodes and pure rGO electrodes.

Sample	Largest Pore Distribution (cc/nm/g)		Pore Diameter (nm)	
	Adsorpsi	Desorpsi	Adsorpsi	Desorpsi
Activated Carbon/rGO composite electrode	0.094835	0.090186	3.0474	3.393
Pure rGO electrode	0.0072449	0.101369	325746	3.068

Table 1 shows the pore size distribution values of Activated Carbon/rGO composite electrodes and pure rGO electrodes. So that the diameter of the pore size on the Activated Carbon/rGO composite electrode is in the range 3.05-3.39 nm, while the pore size diameter of the pure rGO electrode is in the range 3.07-32.57 nm. BJH data is also indicated by desorption-desorption data with the isotherm curve shown in Figure 2.

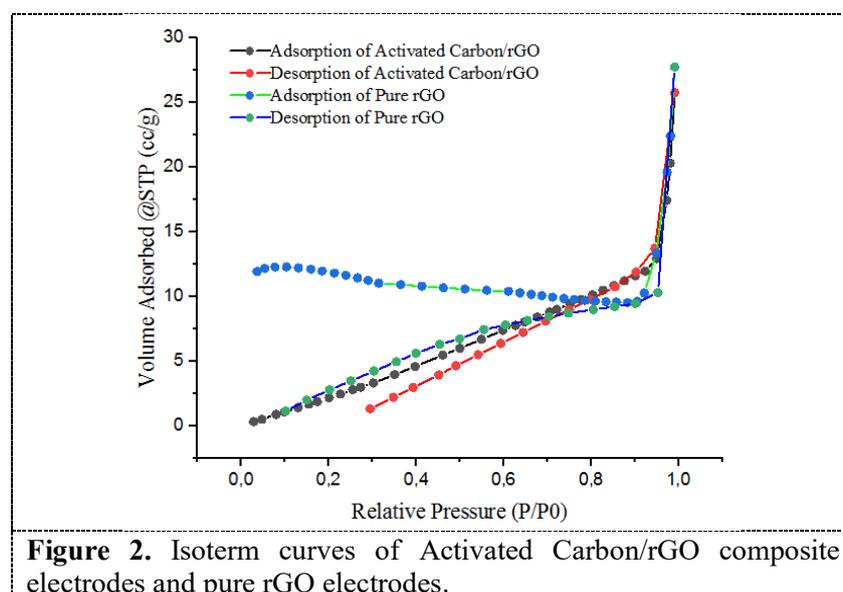


Figure 2 shows the isotherm curves of Activated Carbon composite electrodes and pure rGO electrodes. The curve shows the relationship between relative pressure (P/P_0) at a temperature of 77.3 K and the volume of absorbent gas used ie nitrogen (N_2). The two isotherms curves show the adsorption-desorption process in each sample forming the histerisis loop. The isotherm curve in both samples showed an IV type adsorption-desorption curve type which was a mesoporous characteristic ($2 \text{ nm} < d < 50 \text{ nm}$) with a hysteresis loop type (H1) showing a cylindrical pore shape [14]. On both curves shows the different end of the isotherm curve that is the tip of the open curve indicating that the nitrogen gas absorbed during the adsorption process is retained within the pore so that the volume of the absorbed gas is greater than the volume of gas released at the time of desorption. The curve also forms a hysteresis loop that is for the Activated Carbon/rGO composite electrode formed at a relative pressure range (0.85-0.98 P/P_0) while on the pure rGO electrode at a relative pressure range (0.92-0.98 P/P_0) so that with the formation of the hysteresis loop on the curve shows that the presence of nitrogen gas particles trapped in the pores. Furthermore, the pore size and surface area of each sample are shown in Table 2.

Table 2. Pore Size Value and Surface Area of Activated Carbon sample from coconut shell ^[11], reduced Graphene Oxide (rGO) from coconut shell and Activated Carbon /rGO composite electrode.

Material	Pore Size (nm)	Surface area (m^2/g)
Activated Carbon ^[11]	7.94 – 31.43	2193.47 – 10784.78
reduced Graphene Oxide (rGO)	3.07 – 32.58	15.82 – 84.72
Composite electrode	3.05 – 3.40	102.01 – 10744

The Activated Carbon/rGO composite electrode obtained pore size of 3.05 nm and a surface area of 102.01 m^2/g while in desorption with a pore size of 3.40 nm and a surface area of 107.44 m^2/g . Furthermore, the pure rGO electrode obtained adsorption data with a pore size of 32.58 nm and a surface area of 15.82 m^2/g while in desorption with a pore size of 3.07 nm and a surface area of 84.72 m^2/g . So the pore size of the Activated Carbon/rGO composite electrode is in the range 3.05-3.40 nm while the surface area is in the range 102.02-107.44 m^2/g . Furthermore, the pore size of rGO is in the range of 3.05-32.58 nm while its surface area is in the range 15.82-84.72 m^2/g . Furthermore, the Activated Carbon material synthesized from the coconut shell (*Cocos nucifera*) was characterized by BET in a previous study [5] resulting in BET characteristics using the BJH (*Barret-Joyner-Hallenda*) method the pore size of coconut shell Activated Carbon ranged from 7.94-31.43 nm. While the surface area is in the range 2193.47-10784.78 m^2/g . Activated Carbon and rGO from coconut shell (*Cocos nucifera*) are the basic ingredients of Activated Carbon/rGO composite electrodes, pore size and surface area are known. It shows that the pore surface area of Activated Carbon is bigger than the composite material that is rGO. The large pore surface area is due to the smaller pore size. A large pore surface area will allow maximum absorption of ions. So that it can allow the absorption of ions on the composite electrode does not last maximally because some of the pores of Activated Carbon are covered by rGO which has a large porous size. However, the amount of composition of rGO used is only 10 wt% compared to the amount of Activated Carbon which is more than 90 wt%. So that the composition of rGO that is not too much causes the pore surface area of the electrode not to be covered. It is therefore necessary to use appropriate mass compositions in the synthesis of composite electrodes. By testing BET on Activated Carbon/rGO composite electrodes can show the ability of the electrode in carrying out its function as a supercapacitor electrode which is the process of ion absorption and storage. If the BET test is given absorbate in the form of nitrogen gas (N_2) electrode capable of absorbing nitrogen gas it can be applied to the CV test where the electrode is able to absorb the ion and has good capacitance. In the process of transfer and storage of ions required the right pore size so that it will maximize its electrochemical performance. Furthermore, to determine the electrochemical performance of the electrode, a cyclic voltammetry test is needed to determine the specific capacitance value of an electrode.

A cyclic voltammetry test is used to determine the specific capacitance values held by the electrode. The result of cyclic voltammetry characterization in this research is a voltammogram curve with curve

component on the vertical axis representing Current (A) and on horizontal axis representing Potential (V) shown in Figure 3.

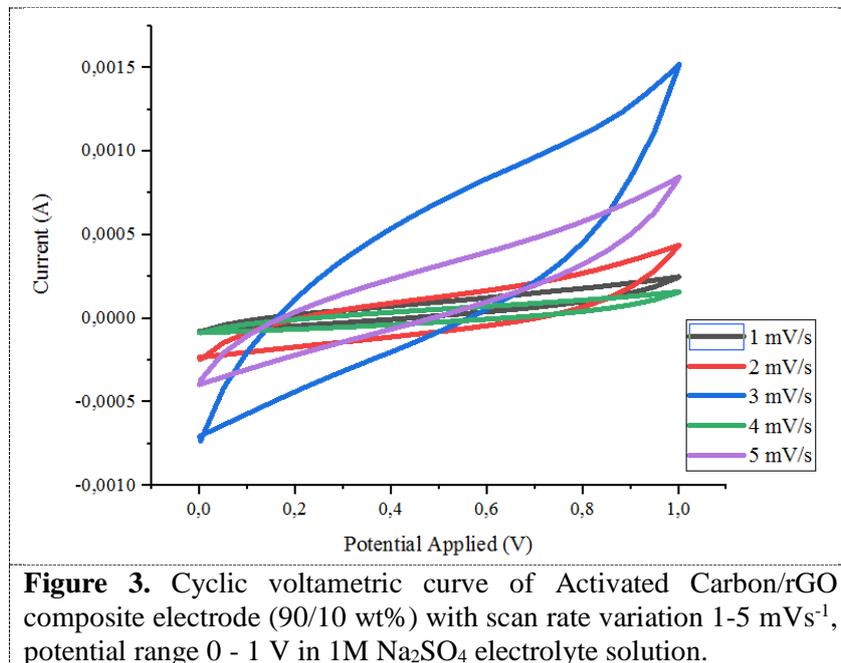


Figure 3 shows the voltammogram curve of the Activated Carbon/rGO composite electrode with a different scan rate variation of 1-5 mVs⁻¹. The potential range of use is at 0-1 V with Sodium Sulfate (Na₂SO₄) 1M electrolyte solution. The voltammogram curve is a reversible curve on each scan rate. The voltammogram curve shows the process of charging and discharging the current on the electrode, where the upper curve is the process of charging current and the lower curve is the process of the release of the current. The voltammogram curve or CV curve shows that there is no significant change in the shape of the curve. This is indicated by the shape of a curve that is still similar to the increase in scan rate but the extent of the curve changes gradually even though the shape tends to be stable. On the CV curve with the smallest scan rate of 1 mVs⁻¹ shows the existence of a more stable storage and release process with a curve shape that almost resembles a horizontal straight line at the top and that of the curve. In contrast to other CV curves where incoming and outgoing currents experience a high or more unstable surge in change. This is evidenced by the shape of the CV curve that rises almost vertically up at the top and jumps down at the bottom of the curve. The CV curve that tends to have a horizontal line or resembles a rectangular shape is an ideal CV curve. The shape of a rectangular-like curve shows that the charge storage process is stable where the density of the current is stable as the potential difference increases. The curve is a curve that approaches the ideal curve conditions on the capacitor [15]. The capacitance value of the Activated Carbon/rGO composite electrode has different values with different use of scan rate values. The result of calculation of specific capacitance value is shown with graph in Figure 4.

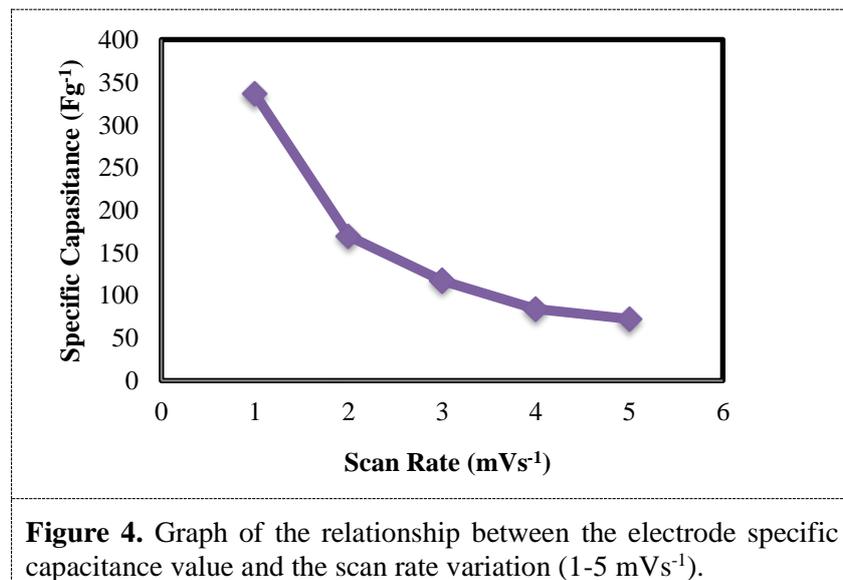


Figure 4 shows a graph of the relationship between the specific capacitance value and the scan rate variation in which the graph decreases the specific capacitance value along with the increase in the scan rate. The specific capacitance value of Activated Carbon/rGO composite electrodes at a scan rate of 1-5 mVs⁻¹ in sequence is 336 Fg⁻¹, 169.5 Fg⁻¹, 117.33 Fg⁻¹, 83.92 Fg⁻¹, and 72.33 Fg⁻¹. From the results of the calculation of the specific capacitance value (C_s) of Activated Carbon/rGO composite electrodes using the Matlab application, the best capacitance value is obtained at the scan rate of 1 mVs⁻¹ by 336 Fg⁻¹. At a scan rate of 2 mVs⁻¹ there was a drastically decreased value of specific capacitance ie from 336 Fg⁻¹ to 169.5 Fg⁻¹. Furthermore, in the scan rate of 3-5 mVs⁻¹ decreases the specific capacitance values slowly. The value of the specific capacitance of the composite electrode decreases with the increase of scan rate used in the CV test. This decrease can be caused because the scan rate value is inversely proportional to the specific capacitance value (C_s). So that the specific capacitance value will decrease along with the increase in scan rate value [5].

In the CV test occurs the process of reduction and oxidation on the surface of the electrode which will then produce measurable electric current at a certain potential. The use of the smallest scan rate in this study is 1 mVs⁻¹ where during the electrolyte storage process generated large cathodic currents to allow for more stored loads. But then with the added value of scan rate, the resulting current is unstable with increasing potential, so it can cause the process of reduction and oxidation is fast. At the time of the reduction process at the electrode poles (charging) occurs the process of adsorption and desorption of ions on the electrolyte so that the formation of a double layer on the electrode and electrolyte [2,16]. At a larger scan rate it can be possible to reduce the process to the smaller cathodic poles, ie electrons that can be bound only by a small amount of material, so that the reduction process takes place quickly and the electrons cannot be completely stored by the electrode poles. The higher the scan rate used, the specific capacitance value will decrease due to the limitations of ion charge transfer on the electrode if high current density [2]. Furthermore, when the process of charging the voltage on the electrode, when the scan rate is given the smaller the flow of the voltage will enter slowly so that it can through the electrode surface to the inside. While if the scan rate is large then the voltage flow can only pass through the outer surface of the electrode so that no charging process is complete. So that this research has succeeded in synthesizing natural Activated Carbon/rGO composite electrodes which are coconut shell (*Cocos nucifera*) with good specific capacitance.

5. Conclusion

The Activated Carbon/rGO composite electrode and pure rGO electrode have different pore size and pore surface area. The pore size of the Activated Carbon/rGO composite electrode is in the range of 3.05-3.40 nm and the pore size of the pure rGO electrode is in the range 3.07-32.58 nm, which was a mesoporous characteristic. So both electrode materials are materials with mesoponic characteristics. The

value of the specific capacitance of the best Activated Carbon/rGO electrode is obtained from the Cyclic Voltammetry (CV) test on the use of 1 mVs^{-1} scan rate with 90 wt% Activated Carbon composition and 10 wt% rGO of 336 Fg^{-1} , while the specific capacitance value of the electrode, the smallest is present at the 5 mVs^{-1} scan rate of 72.33 Fg^{-1} . So the use of a small scan rate causes the capacitance value owned by the electrode to be better [2].

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