

# Preparation and Characterization of Activated Carbon Based on Coconut Shell for Supercapacitor

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# Abstract

The development of activated carbon from coconut shell charcoal has been investigated by using physical and chemistry method to determine the influence of activation parameters in term of temperature, Nitrogen gas pressure and time period on the pore structure of the activated carbon. The coconut shell charcoal process at the temperature of about 800°C for 5 hours. The charcoal was activated by pyrolysis process at various temperature (600,700 and 800°C) for 2 hours, The results showed that the pores size were reduced and distributed uniformly as the activation parameters are increased. Pore size and surface area Characterized by BET method, and performance supercapacitor characterized by cyclic voltammetry.

Keywords: Coconut shell charcoal; Activation process; Carbon; Pore distribution; Supercapacitor.

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#### 1. Introduction

Activated carbon can be produced from different raw carbon resources like lignite, peat, coal, and biomass resources such as wood, sawdust, and coconut shells [1]. Activated carbon is an amorphous carbon from flat plates prepared by C atoms covalently bonded in a hexagonal lattice with one atom C flat on each the angle that the surface area ranging from  $300 \text{ m}^2/\text{g}$  to  $3500 \text{ m}^2/\text{g}$  and is related to the internal pore structure that has the properties as an adsorbent. There are two types of activation processes to produce activated carbon from carbonaceous materials, i.e. chemical and physical activation processes [2].

Physical activation procedure involves a two-step process, i.e. carbonization, followed by activation using steam, oxygen or carbon dioxide as an activating agent [3]. The activation process is a treatment for people who aim to enlarge the pore that is by breaking the bonds of hydrocarbons or oxidizing surface molecules that change the nature of charcoal, both physics and chemistry, the surface area increase in size and influence on the adsorption capacity. In general, activated carbon can be in the activation of the two ways, chemical activation with alkaline metal hydroxide, carbonate salts, chlorides, sulfates, phosphates of alkaline earth metals and especially  $ZnCl_2$ ,  $CaCl_2$ , inorganic acids such as  $H_2SO_4$  and  $H_3PO_4$  and activation physics which is the process of breaking the carbon chains of the organic compound help heat at a temperature of  $800^{\circ}C$  to  $900^{\circ}C$  [4].

Influence the activation process is time, temperature, the particle size, the ratio of activator and type of activator which in this case will affect the absorption of active charcoal activated carbon can be prepared from any organic material carbon and preferably with low content in inorganic matter[5]. The processes more commonly used to prepare materials are physical and chemical activation. For physical activation (also called thermal activation) the char obtained by carbonization of the precursor is partially gasified by an oxidant atmosphere (usually steam or carbon dioxide) at temperatures in.

The impregnated material is then carbonized under an inert atmosphere and, finally, the carbonized material is washed to remove the chemical and to leave a porous structure available. The main differences are that carbonization and activation are usually independent steps during physical activation, where as the steps cannot be separated during chemical activation the chemical acts on the material being carbonized) [6]. Carbonization is a process for converting organic material into charcoal, the carbonization process will release a flammable substance such as CO,  $CH_4$ ,  $H_2$ , formaldehyde, methane, formik and acetil acid and unburned substances such as  $CO_2$ ,  $H_2O$ , and liquid tar. Gasses that are released in this process has a high calorific value and can be used to meet the needs of heat in the process of carbonization. The activation process is important addition to raw materials used [7].

What is mean by activation is a treatment against charcoal aims to enlarge the pore that is by breaking the bonds of the molecules oxidize hydrocarbons or carbon surface so change the nature, both physics and chemistry, the surface area increase in size and influence on the adsorption capacity. A process that involves the selective oxidation of the raw material with air, are also used both for the manufacture of activated charcoal as well as an absorbent. Carbonized raw material at a temperature of 400-800°C to eliminate substances that are volatile. Then oxidized with gas at 600-1000°C to made pores and surface area[8].

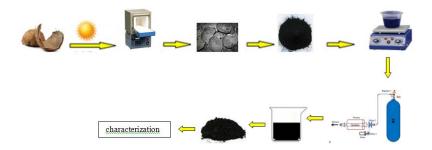


Figure 1: Schematic preparation activated carbon

In this study, carbonization was used to produce coconut shell activated carbon using KOH as an activating agent. The properties of the activated material such as surface area, pore volume and pore size.

The raw material is mixed with certain chemicals, then created on. Next on the molded into bars and dried. Activation is done at temperatures of 100 °C. The activated charcoal, washed with water and then dried at a temperature of 300°C. With chemical processes, the raw material can be carbonized in advance, then mixed with chemicals. At this chemical activation results from carbonization charcoal soaked in a solution of activation before heated. In the chemical activation process, charcoal soaked in the solution for 24 hours then drained and heated at 600-900°C for to 2 hours [9].

Supercapacitors have many advantages compared with other energy storage devices such as batteries. From the technical side, supercapacitors have a relatively large number of cycles (> 100,000 cycles), high energy density a great ability to save energy, the principle is simple and convenient construction [10]. In terms of user-friendliness, supercapacitors improve security because of no corrosive materials and fewer toxic materials. Currently, the electrode material of commercial supercapacitors use a porous carbon made from natural materials, namely coconut shell. The porous carbon can be prepared by carbonization of natural materials such as coal, coconut shells, corn stalks, wood, bamboo, paper waste [11]. Porous carbon potential to be used as supercapacitors electrode material because it has the potential for high energy density, pore good accessibility and relatively low manufacturing costs. To increase the capacitance of supercapacitor energy can be done by preparing porous carbon material which has mesoporous fraction and high surface area. The material used for the manufacture of a supercapacitors electrode including graphene, carbon nanotubes, carbon aerogel, a porous carbon and minerals-carbon composites [12].

The supercapacitor was made of symmetric electrode. The capacitance was determined using Eq. 1:

$$C = \frac{dq}{dv}$$
(1)

Current is constant, there for:

$$C = I \frac{\mathrm{d}t}{\mathrm{d}v} \tag{2}$$

Total capacitance obtained by a combination of two equivalents of the electrodes arranged in series. Therefore, the equation for the capacitance C for 2 cells is.

# 2. Experimental

#### **Materials and Methods**

#### 2.1. Preparation of activated carbon

Coconut shell as a precursor material. Clean the coconut shell from other materials such as coconut fiber or soil. Sun dry it for 2–3 days. The dried coconut shell was then kept in furnace at 800°C for 5 h. as an activating agent is one of the most effective agents employed for organic materials. KOH is mostly preferred over other activating agents because it causes more localized reactions with the carbon precursor and is more effective for highly ordered materials. Chemical activation has been widely used to obtain porous activated carbons with very high surface areas. The raw material was ground and sieved into sizes of less than 200 mesh to ensure that they were thoroughly mixed with an activating agent during the impregnation process. The particles of coconut shell mix to KOH (1:3). The impregnated sample was heated using an oven at 90°C for 2 hours. The heated process occurred in the presence of nitrogen gas (100 ml min-1) for 2 hours. The pure nitrogen gas used was 98%. The sample was washed using aquades to remove excess KOH until pH 6.5–7.0. The sample was then dried in an oven a 100°C for 2 hours.

#### 2.2. Preparation of Electrodes

# Manufacture of sheet electrodes

PVDF + DMAC stirrer at a temperature of 80 °C for 1 hour and then added (Activated carbon, super-P) and stirred for 3 hours until homogeneous. After that, coating on the aluminum sheet by Dr. Blade method. Dry on open air (make sure the sheet material does not crack). And then sheet carbons were oven at a temperature of 80 °C for 1 hour).

#### 2.3. Assembly cell supercapacitors

The capacitor cells were constructed with an electrolyte-impregnated polypropylene separator sandwiched between two electrodes. The electrodes were prewetted with electrolyte before use. The supercapacitors performance was analyzed by the Cyclic Voltammetry instrument test.

#### 3. Result and discussion

The surface area and pore volume increased proportionally to the KOH/carbon weight ratio up to its value of about 3. A surface area greater than 1000  $m^2/g$  was obtained at a KOH carbon ratio of about 3.The reaction of KOH and surface carbon (C) occurring during activation process is as follows [13]:

$$4 \text{ KOH} + \text{C} \implies \text{K}_2 \text{CO}_3 + \text{K}_2 \text{O} + 2\text{H}2 \tag{3}$$

$$K_2CO_3 + 2C \implies 2K + 3CO$$
 (4)

$$K_2O + C \qquad = > C - O - K + K \tag{5}$$

$$C-O-K + H_2O \implies C-O-H + KOH$$
(6)

The reaction on surface of carbon could be explained by the following equation :

$$2KOH \implies K_2O + H_2O (dehydration)$$
(7)

$$C + H_2O = H_2 + CO$$
 (water gas reaction) (8)

$$CO + H_2O = H_2 + CO_2$$
 (Water gas shift reaction) (9)

$$K_2O + CO_2 \implies K_2CO_3$$
 (carbonate formation) (10)

The final product of activation in the presence of KOH during the preparation of activated carbon is  $K_2CO_3$ . K metal was produced during the activation process at < 700°C is show in following equation :

$$K_{2}O + H2 = > 2K + H2O \text{ (reduction by hydrogen)}$$
(11)  
$$K_{2}O + C = > 2K + CO \text{ (reduction by carbon)}$$
(12)

A number of experiments were carried out to study the effect of the parameters on activation. The major parameters that affect the quality of activated charcoal were temperature ratio. Activation increased with an increase in temperature. At low and high temperatures activation is low porosity [14]. From result BET analysis we found that the surface area and pore size decreasing when temperature increasing.

Sample			Pore	Volume	Pore	Size
Code	Temperature	Surface area $m^2/g$	(cc/g)		(Angstrom)	
А	800	1050	0,262		13,72	
В	700	1469	0,457		24,51	
С	600	1089	0,173		14,36	

Table 1: Parameter of the sample Activated Carbon

The higher the temperature oven, the more pores are open so that the extent of increases, this will lead to many components of substances such as water content and volatile are still entrapped going out, resulting in severe activated charcoal will shrink quite a lot at a temperature of 800°C to about 70% of them. Activated carbon adsorption capacity for nitrogen has a correlation with a surface area of activated carbon. The greater the surface area ability to adsorb or solutes. The greater absorption as the temperature rises, this means that the quality

better than activated charcoal [15].

Figure 2 show the pore distribution majority 2-3 nm, its means that pore are form at high fase mesoporous.

The conductivity of sample activated carbon B is the highest, demonstrating that the reactivation is favorable to prepare the activated with higher conductivity

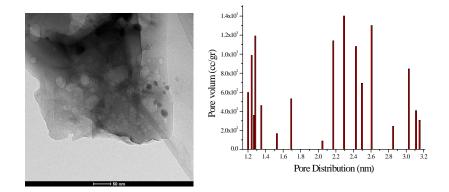


Figure 2: TEM Activated Carbon at 700°C

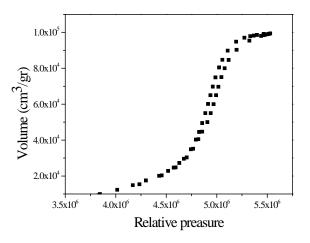


Figure 3: Isotherm fisisorption and pore size distribution

Based on the above results can be analyzed the effect of temperature on the activation of carbon produced. It appears that the increase in activation temperature can increase the surface area and pore volume. This is possible because of the increased reactivity of the carbon atom K. Effects of temperature between the carbon and the KOH activation have been described in the literature [16].

High mesoporous rate is suitable for electrolyte ion to transport inside the porous structure of the activated carbon. More inner-surfaces of pores inside the AC can be wetted by the electrolyte. Therefore, the conductivity of activated carbon electrode is enhanced. When an electrode (electronic conductor) comes into contact with an

electrolyte (ionic conductor), it shows some potential and attracts ions with the opposite sign, forming an "electrical double-layer" at the electrode/electrolyte interface. The specific capacitances of the three Activated Carbon electrodes are 86 F/g (A), 110 F/g (B) and 98 F/g (C)

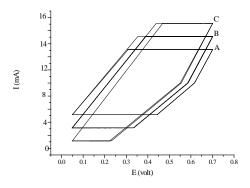


Figure 4: Cyclic voltammogram of activated carbon at scan rate 10mv/s

# 4. Conclusion

Activated carbon with high mesoporous ratio were prepared by chemical activation followed by physical activation based on coconut shell carbon. Their capacitive behaviors and conductive characteristics were investigated based on CV measurements. It was found that the capacitive behavior was improved as the mesoporous ratio increased. The reason is believed to lie in the difference among their mesoporous ratio and conductivity. High mesoporous ratio is favorable for the ion diffusion in the meso/micro-pores. The process enhances the accessibility of the micropore surface area and the conductivity of activated carbon electrode material. In conclusion, activated carbon with higher mesopore ratio can offer a great potential in supercapacitors.

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#### References

- Ioannidou, O., Zabaniotou, A.,(2006), "Agricultural residues as precursors for activated carbon production" a review. Renewable and Sustainable Energy Reviews 11 (9), 1966–2005
- [2]. YuenFK & haMeed BH.(2009), "Recent developments in the preparation and regeneration of activated carbons by microwaves", Advance in Colloid and Interface Science 149: 19–27.
- [3]. NciBiMC, Jeanne-roSeV, MahJouBB, Jean-MariuSc, laMBertJ, ehrhardtJJ, Berciony, SeFFenM & gaSPardS, (2009), "Preparation and characterization of raw chars and physically activated carbons derived from marine Posidonia oceanic (L.) fibres", Journal of Hazardous Material 165: 240–249
- [4]. H.Marsh, F. Rodríguez-Reinoso, (2006)," Activated Carbon", Elsevier, United Kingdom

- [5]. G. Afrane, O.-W. Achaw, (2008), "Bioresource", Technol. 99
- [6]. M. Molina-Sabio, F. Rodríguez-Reinoso, (2004), "Colloids Surf", A 241
- [7]. J.M. Ramos-Fernández, M. Martínez-Escandell, F. Rodríguez-Reinoso, (2008) Carbon 46: 384.
- [8]. An, K. H., Kim, W. S., Park, Y. S., Choi, Y. C., Lee, S. M., Chung, D. C., Bae, D. J., Lim, S, "Electrode Material for Supercapacitors", Carbon, vol.45, pp. 160-165.
- [9]. Miller, J.R. and Simon, P., (2008), "Supercapacitors: Fundamentals of Electrochemical Nanohorn/ Nanotube Composite", ACS Nano, Vol. 5, No. 2, pp. 811-819. Proceedings of the 2002 NSF Design, Service and Manufacturing Grantees and Research.
- [10]. Hata, K., (2011), "Supercapacitor Electrodes from Single- Walled Carbon", High-Power
- [11]. Y. Ji,T.Li, L.Zhu,X.Wang,andQ. Lin, (2007) "Preparation of activated carbons by microwave heating KOH activation", Applied Surface Science,vol.254,no. 2, pp.506–512
- [12]. BaBelK & JurewiczK, (2004), "KOH activated carbon fabrics as supercapacitor material", Journal of Physical and Chemical Solids 65: 275–280.
- [13]. Liw, PengJ, zhangl, yangK, Xiah, zhangS & guoSH, (2009), "Preparation of activated carbon from coconut shell chars in pilot-scale microwave heating equipment at 60 kW", Waste Management 29: 756–760.
- [14]. Liw, zhangLB, PengJH, lin& zhu XY,(2008), "Preparation of high surface area activated carbons from tobacco stems with K<sub>2</sub>CO<sub>3</sub> activation using microwave radiation", Industrial Crops and Products 27: 341–347.
- [15]. NciBiMC, Jeanne-roSeV, MahJouBB, Jean-MariuSc, laMBertJ, ehrhardtJJ, Berciony, SeFFenM & gaSPardS, (2009), "Preparation and characterization of raw chars and physically activated carbons derived from marine Posidonia oceanic (L.) fibres", Journal of Hazardous Material 165: 240–249
- [16]. Zhang, F.M.H., Chen, J., Li, G.D., Zhang, Y., and Chen, J.S., (2008), "Preparation and Gas Storage of High Surface Area Microporous Carbon Derived from Biomass Source Cornstalks", Bioresource Technology, vol. 99, pp. 4803-4808