

# DESIGNING AND DEVELOPING RECHARGEABLE ALUMINIUM-ION BATTERY USING GRAPHITE COATED ACTIVATED CHARCOAL CORNCOB AS CATHODE MATERIAL

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## ABSTRACT

One of the renewable energy storage systems that can be used today is the aluminum ion battery. In this study, aluminum foil was used as anode, polyethylene polypropylene (PE/PP) as separator, electrolyte from  $AlCl_3/[EMIm]Cl$  and graphite coated corncob, an activated charcoal, as cathode. Coating method of cathode materials was done by mixing both graphite and activated charcoal with varied composition 1:0.5, 1:1, 1:1.5, and 1:3. The coating process began by mixing the graphite and corncob with ethanol as a solvent for six hours, then heating in an oven at 80 °C for three days, gradual drying in a furnace at 350 °C for five hours and sintering at 600 °C for six hours. From this research, SEM results showed that carbon particles were evenly distributed, with spherical particles. The spherical shape was the main requirement of carbon formation in order to produce high energy. Based on the results, battery potential was 2.54 V with average of optimal capacity at a ratio of graphite and corncob activated charcoal 1:1.5 was 83.067 mAh/g. The highest efficiency was also at a ratio of 1:1.5 of 97.20%, because at this ratio, there was an increasing in percentage of element C 91.74%, greater than the percentage of element C on the other three cathode samples.

## ABSTRAK

Salah satu sistem penyimpan energi terbarukan yang bisa digunakan saat ini adalah baterai ion aluminium. Pada penelitian ini digunakan aluminium foil sebagai anoda, polyethylene polypropylene (PE/PP) sebagai separator, elektrolit menggunakan  $AlCl_3/[EMIm]Cl$  dan grafit terlapisi arang aktif tongkol jagung sebagai bahan katoda. Metode pelapisan bahan katoda dilakukan dengan mencampurkan grafit dan arang aktif dengan variasi komposisi 1:0,5, 1:1,1:1,5 dan 1:3. Proses pelapisan diawali dengan pencampuran grafit dan arang aktif tongkol jagung dengan ethanol sebagai pelarut selama enam jam kemudian pemanasan di oven pada suhu 80oC selama tiga hari, pengeringan bertahap di furnace pada suhu 350oC selama lima jam dan sintering pada suhu 600oC selama enam jam. Dari penelitian ini didapatkan hasil SEM menunjukkan bahwa partikel karbon terdistribusi merata, dengan bentuk partikel bulat (sphere). Sampel berbentuk bulat atau sphere merupakan syarat utama pembentukan karbon supaya dapat menghasilkan energi tinggi. Berdasarkan hasil uji baterai diperoleh potensial sebesar 2,54 Volt dengan rata-rata kapasitas optimal terjadi pada rasio grafit dan arang aktif tongkol jagung 1:1,5 sebesar 83,067 mAh/g. Efisiensi tertinggi juga terjadi pada rasio 1:1,5 sebesar 97,20%. Hal ini karena pada rasio 1:1,5 terjadi peningkatan persentase unsur C yakni 91.74% lebih besar dari persentase unsur C pada tiga sampel katoda yang lainnya.

**Keywords:** Aluminum Ion Battery; Corncob Activated Charcoal; Coating; Cathode; Graphite

## INTRODUCTION

Battery is one of the need for human in energy storage. The development of renewable energy for power generation and transportation requires *energy storage* in the form of

batteries.

Battery is electrochemical cells that produces a constant voltage from the electrochemical reaction. Battery is an electronic device that converts chemical energy into electrical energy. The battery has two electrodes in which a chemical reaction will take place by inoculating an electron. Both the anode and cathode electrodes are connected with a solution called

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the electrolyte where ion could move in it. The movement of these electrons will produce an electric current as a source of energy for electronic equipment (Sagir & Mohd, 2011).

On an aluminum-based battery, the anode is one of the most important components. Theoretically, the generated voltage depends on the metal type of its anode. This difference is based on the standard potential energy value of each metal (Vincenzo & Benedetko, 2014).

Aluminum acts as an anode on the battery, so this battery is named as an aluminum ion battery. The aluminum will oxidize resulting and electrons. Then, this electron will move towards the cathode. The movement of electrons to the cathode will produce electrical energy (Modesto & Julie, 2007). The aluminum battery is described as a battery that has high enough potential. This battery can be applied in various fields of application including in the military field. This is because aluminum batteries have a pretty good advantage such as light and rechargeable power (Rao et al., 1992)

One type of cathode used in aluminum-based batteries is graphite or three-dimensional grafit foam (Meng-Chang et al., 2015). In addition, other types of cathodes used in aluminum-based battery systems consist of three main components: porous carbon, catalysts and binder polymers (Jang et al., 2011).

This study used graphite coated with porous carbon contained in corncob activated charcoal as cathode. The porous carbon based on the structure pattern is an *amorphous* carbon material composed largely of free carbon and has an inner surface, thus having a high absorptive capacity (Alfathoni G., 2002). Activated carbon has a good absorption capacity to the solution and gas, activated carbon is one of the most frequently used adsorbents in the adsorption process. This is because the activated carbon has better absorption and surface area than other adsorbents (Walas, 1990). The nature of the activated carbon may be used as an electrode or as a gas absorbent medium to perform electrochemical processes on aluminum-based batteries (Yugang, 2013).

In addition, the use of activated charcoal as a coating of graphite material in anode making due to activated charcoal is one of the leading raw materials to increase storage capacity of a battery. So in this study, cathode was prepared with carbon source from amorphous carbon-coated graphite from corncob activated charcoal so that it can improve storage performance of aluminum ion battery.

The purpose of this study are: first, to know the effect of variations in graphite composition coated with corncob activated charcoal on microstructure of cathode material; second, to determine the effect of variations in graphite composition coated by corncob activated charcoal on quality of graphite coating; and the last, to know the influence of variation in graphite composition coated by corncob activated charcoal on the performance of aluminum ion battery (battery capacity and battery efficiency)

## METHOD

This research was an experimental research conducted at Analytical Technique Laboratory of Mataram University and Battery Laboratory of Physics Research Center (PRC) – Indonesian Institute of Science (IIS) Serpong Tangerang Selatan. In this research, there were several stages of activity or workmanship, namely cathode material synthesis, material characterization, slurry making, sheet making, calendaring and cutting Sheets, drafting to battery life, and battery performance testing.

### Synthesis of Cathode Material

The first step in this research was the preparation of cathode materials for aluminum ion batteries. The synthesis process of graphite material was done by mixing method (graphite and corncob activated charcoal ) with 50 mL ethanol on a hot plate at a temperature of 80°C hot plate and spun at 200 rpm for six hours. Afterwards, the sample was stored in an oven 80°C to remove the water content for three days until gel deposits was formed in the above prior to heat at 350° C for five hours called degradation process. Degradation process is a reaction of chemical change or decomposition of a compound or molecule into a simpler compound or molecule gradually. Heating for the release of hidrogen, so only remaining carbon in the sample. Next, resulting sample was crushed and reheated at 600° C sintering temperature for four hours, this was called the carbonization process. Carbonization is a process whereby the oxygen and hydrogen elements are removed from the carbon and will produce a carbon framework having a specific structure.

### Material Characterization

The samples of the material have been made and characterized by using Scanning Electron Microscopy (SEM) Hitachi SU3500 brand with tungsten as the source of electron

to observe the morphology and particle size.

### Aluminum Ion Battery Making

Battery making was done in several stages. The first stage, slurry making, prepared graphite coated corncob activated charcoal sample with variation of composition (1:0.5, 1:1, 1:1.5, 1:3), PVDF (Polyvinylidene Fluoride), AB (Acetyllene Black), and DMAC (NN Dimethyl Acetamid) according to the composition of 85%:10%:5% and DMAC  $\pm$  12 mL. PVDF powder acted as a binder, AB acted as an additive and DMAC acted as a solvent, then the hot plate to be used to make the slurry set with a temperature of 70°C with rotation of 150 rpm. First, mixed DMAC with PVDF at hot plate with magnetic stirrer inside beaker glass until clear then put AB into DMAC and PVDF solution slowly until well mixed. After being well mixed, inserted graphite coated corncob activated charcoal slowly and left mixed over hot plate until the desired slurry was formed  $\pm$  1.5 hours.

The second stage, sheet making, put Cu-foil over the doctor blade and vacuumed it, then Cu-foil was cleaned by acetone. The thickness of the doctor blade was measured with a thickness gauge of 0.2 mm and then poured slurry on Cu-foil little by little and doctor blade was run at speed of  $\pm$  6-7 rpm until Cu-foil was perfectly coated by slurry. After finished, then graphite anode sheet was dried on a dry box at temperature of 80°C until dried ( $\pm$  1 hour).

The third stage, calendering, which include part of the sheet making process. Calendering was the process of pressing the sheets using a roll consisting of two tubes to ensure that the sheets did not fall out and were already sealed.

The fourth stage, *cutting*, the process of cutting the sheets for further used as an aluminum ion battery cathode. This cut was formed a circle with a diameter of 1.5 cm.

The last stage, battery assembly, prepared tools and materials needed then arranged in sequence from the base (can), spaser, anode, separator, and cathode. The used cathode was graphite coated with corncob activated charcoal and separator in the form of polyethylene polypropylene (PE/PP). Dropleted electrolyte  $\text{AlCl}_3$ /[EMIm]Cl 2 M until the entire surface was exposed by electrolyte liquid, then placed spaser above it, next wave spring. Finally sealed with a seal and sealing chassing perfectly to cover it tightly. Left for one day before tested using battery analyzer (MTI)



**Figure 1.** Design of aluminum ion battery (coin cell).

### Battery Testing Process

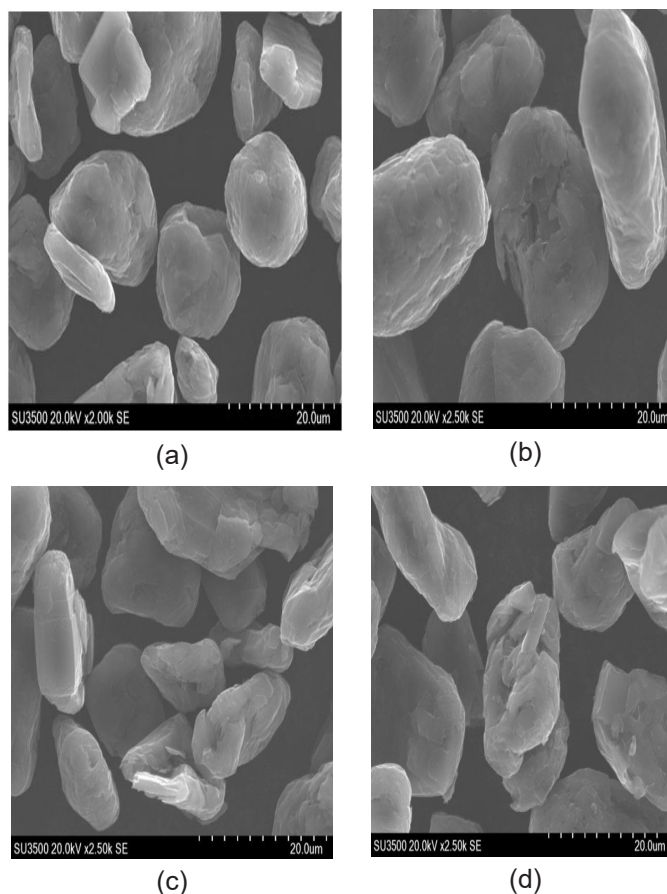
Battery testing was done using a battery analyzer (MTI) with the first step, put the battery in the terminal on the battery analyzer (there were 8 channels). Then the BST8 software application (battery system test 8) was opened and ensured the software was connected to the battery analyzer. Next the battery parameters was set by right click and select startup. The parameters set included: constant current discharge/discharge current limit condition, constant current charge/current charging limit condition, Constant voltage charge/charging limit condition, Cycle/number of cycles and rest / break time. Then pressed Ok, the battery analysis process run to completion according to how many cycles were set.

## RESULTS AND DISCUSSION

The graphite synthesis of corncob activated carbonated was carried out with four variations of composition ie 1:0.5, 1:1, 1:1.5, and 1:3 used as cathode materials on aluminum ion batteries. This research was conducted by several stages. The first stage was making of cathode from graphite coated corncob activated charcoal then identify its micro structure with SEM/EDX. The next step was the preparation of batteries include making slurry, sheet making, until battery assembly. Electrochemical characterization using MTI.

### Characterization of microstructure using SEM (Scanning Electron Microscopy)

The microstructures of graphite particles were characterized using Hitachi SU3500 SEM with *Energy Dispersive X-ray* (EDX). The results of SEM of the sample were shown in Figure 2.



**Figure 2.** The result of SEM (a) graphite coated activated charcoal 1:0.5, (b) 1:1, (c) 1:1.5, and (d) 1:3.

Figure 2. shows photos SEM result of pure graphite and graphite coated activated charcoal with a variation of 1:0,5, 1:1, 1:3 composition. Observations were made at 2500 magnification times so that the morphology of the particles could be seen clearly.

**Table 1.** Average grain size of pure graphite samples and variations of graphite composition with activated charcoal .

Sample	Composition	Average grain size ( $\mu\text{m}$ )
A	1:0,5	108,518
B1	1:1	128,74
B2	1: 1,5	93,228
B3	1:3	103,161

Table 1. shows pure graphite with average grain size of 108.518  $\mu\text{m}$  will change after coated with activated charcoal. The smallest grain size is present in the graphite sample which is coated activated charcoal on a 1:1 composition.

**Table 2.** The distribution of carbon (C) and oxygen (O) in pure graphite samples and graphite coated activated charcoal with composition 1:0.5, 1:1, 1:1,5, 1:3.

Sample	Content	Weight%	Atomic%
A	CK	89.45	91.86
	OK	10.55	8.14
B1	CK	91.35	93.36
	OK	8.65	6.64
B2	CK	91.74	93.67
	OK	8.26	6.33
B3	CK	83.35	91.36
	OK	16.5	8.64

The result of SEM test on graphite coated activated charcoal with varied composition in photo scanning with 2500 times magnification. Spherical shape is the main condition of carbon formation for producing a high energy. Composite of metal elements with higher atomic numbers will result in lighter/brighter color than the constituent metal elements with lower atomic numbers.

The largest weight of carbon is shown in the variation of graphite composition with activated charcoal is sample B2 (1:1,5). This shows that in this sample formed the perfect carbonization at the time of *sintering* so that O more released and left only C, otherwise the sample B3 (1:3) has less distribution element C this shows that in this sample has not yet formed a perfect carbonization during *sintering* so that O has not much to detached.

### Aluminum Ion Battery Performance Analysis Battery Capacity

The capacity of an aluminum ion battery with an initial potential of 2.54 V is shown by the following Figure 3.

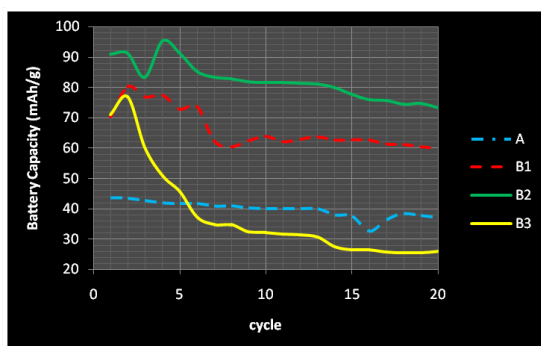


Figure 3 . Battery capacity per cycle.

The average battery capacity of sample A was 39.75 mAh/g, sample B1 was 65.95 mAh/g, sample B was 82.09 mAh/g, and samples B3 was 37.67 mAh/g. Figure 3 shows the greatest capacity occurring in sample B2 with a capacity of 95.37 mAh/g at the 4th cycle and its capacity drops to 73.203 mAh/g at the 20th cycle. This is proportional to the carbon percentage in Table 2. The lowest capacity occurs in the sample B3 of 25,509 mAh/g at the 18th cycle.

### Battery efficiency

Battery efficiency is obtained from capacity test data during *charge* and *discharge process*. The value of battery efficiency was obtained from the comparison of discharge and charge capacity.

Average of battery efficiency in sample A was 96.53%, sample B1 was 68.74%, sample B2 was 97.20%, and sample B3 was 93.37% at 20 cycles. So the battery samples of A, B2, and B3 tend to have longer lifetime than battery sample B1, where battery B1 sample will run out faster in its application.

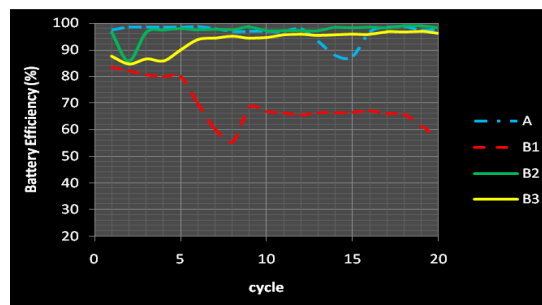


Figure 4 . The efficiency of the battery.

### CONCLUSION

In this study we can conclude that the effect of variation in composition of graphite coated by corncob activated charcoal on the quality of graphite coating is shown by SEM analysis. The coating will change the average grain size and distribute the carbon particles well, with spherical shape.

The capacity of aluminum ion batteries with a potential of 2.54 V shows an increase that is proportional to the increase in the percentage of carbon on the cathode sample. Average of optimal capacity occurred in the ratio of graphite and corncob activated charcoal 1:1.5 of 83.067 mAh/g. The highest efficiency also occurred at a ratio of 1:1.5, i.e. 97.20%.

Further research can be done by using variation of graphite composition with activated charcoal besides corncob active charcoal with the same composition that is 1:0,5, 1:1, 1:1,5 and 1:3. Besides, higher sintering temperatures gives better results.

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