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Effect of Solid Content in Electrochemical Performance of Graphite Anode of Lithium-ion Batteries

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Abstract. Rechargeable batteries have been implemented in most portable electronic devices. Lithium-ion battery (LIB), as the main power source, dominates the mobile device market due to its high energy density, long shelf life, and environmentally friendly operation. In the rechargeable lithium-ion battery, there are four main components, one of which is the anode. The anode material used is commercial graphite. Thus, this study aims to determine the effect of solid content solvents on battery performance. The main discussion in this study is to analyze the effect of solvent variations of N, N Dimethyl Acetamide (DMAC) on the characteristics of the sheet and the difference in solid content of graphite anode sheets on battery performance. Identification of the formed phase was carried out by XRD, reduction and oxidation reactions by cyclic voltammetry test, battery capacity by charge/discharge test, and study of the electrochemical characteristics of the electrode material by electrochemical impedance spectroscopy test. The best anode sheet is produced by mixing 2.5 mL DMAC solvent at a thickness of 0.07 mm with a solid content of 25%. The results of the charge-discharge test showed a specific capacity of 309.33 mAh/g in the first cycle.

Keywords: Lithium-ion Battery, Graphite, Solid Content, Specific Capacity

1 Introduction

Nowadays, rechargeable batteries have been implemented in most of the portable electronic devices. Lithium-ion battery (LIB), as the main power source, dominates the portable device market due to its high energy density, long shelf life and environmentally friendly operation [1-3]. In order to meet market portability, lithium battery development creates a lightweight, thin, flexible and small battery. Meanwhile, the types of batteries that are widely used are still too heavy, thick, stiff and bulky [4].

In this case the lithium battery becomes the object of research with the aim of further improving its properties and characterization. One of the components in the lithium battery cell system is the anode [5-6]. And despite extensive research efforts to find suitable alternatives with

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increased power or energy density, while maintaining excellent cycle stability, graphite is still used in most commercial LIB available today.

In this research, we will make anode sheet with the active material of commercial graphite. The graphite anode sheet is made in a half cell battery in the form of slurry. In addition, the composition of the solvent N,N Dimethyl Acetamide (DMAC) will be varied which is used to make anode sheets. The use of solvents will have a thickening effect on the slurry, which is the result of mixing the composition of the ingredients or is called a slurry. From the variation of solvents and different thicknesses, it will be seen the ratio of solid content that affects the performance of lithium-ion batteries. Then the anode sheet is cut with a diameter of 16 cm, with a polyethylene separator, and $LiPF_6$ as an electrolyte which is then formed into a lithium battery in the form of a coin cell. Powder characteristics test was carried out by XRD. Battery performance tests include cyclic voltammetry, charge/discharge and electrochemical impedance spectroscopy tests. This research is expected to help determine the composition at the time of making slurry so as to produce good solid content and create the right sheet thickness and produce good battery performance.

2 Methods

The manufacture of graphite-based anode sheets for lithium batteries is carried out in several stages. The manufacture of this battery anode sheet consists of three materials, namely graphite powder as an active material in the manufacture of batteries. The second ingredient is PVDF powder as a binder which strengthens the anode of the lithium-ion battery as well as being an adhesive for graphite with addictive substances. The third material is Super-P which is an additive that functions as a conductive carbon to increase the conductivity of the graphite anode. In mixing the three ingredients, a DMAC solvent is needed.

The process of making slurry is carried out with the composition of the weight ratio in a row, namely Graphite: PVDF: Super P (80:10:10). In a sample of 0.5 grams of Graphite, 0.0625 grams of PVDF are needed, 0.0625 grams of Super-P are needed as a solvent for raw materials. Next, fill a 5 mL beaker glass with DMAC and place a magnetic bar inside the beaker glass. Turn on the hot plate HS 65 and place the beaker glass on top of the magnetic stirrer and set the heating temperature to 50°C with a rotation speed of 300 rpm. Add the PVDF material into the glass beaker gradually and wait until it is homogeneous for about 15 minutes then add the next ingredients, namely Super P and Graphite in the same way. After all the ingredients are mixed, the sample is waited for the stirring of the filler material in the matrix for 30 minutes.

The finished slurry is then poured on top of the Cu sheet which is placed on the coating machine. When the coating process is complete, the sheet is oven-dried. Furthermore, in the manufacture of coin cells, the anode sheets are cut with a diameter of 16 mm and then arranged in an orderly manner to form a coin cell and the assembly process is carried out.

In the manufacture of graphite anode sheets, tests will be carried out to determine the characteristics of graphite and the stages of testing are Cyclic Voltammetry, Charge Discharge and Electrochemical Impedance Spectroscopy.

3 Result and Discussion

3.1 Graphite XRD Results

X-ray diffraction (XRD) is an analytical method in crystallography. This is a way to determine the atomic structure of a crystal, but nowadays it can be used to determine much more information, such as cell parameters, distortion, tension and crystal size obtained by XRD [7].

Testing on graphite powder was done using XRD. The test was carried out with X-rays using a wavelength of Cu-Kα (λ=1.54187) which aims to determine the type of phase and the size of the crystalline.

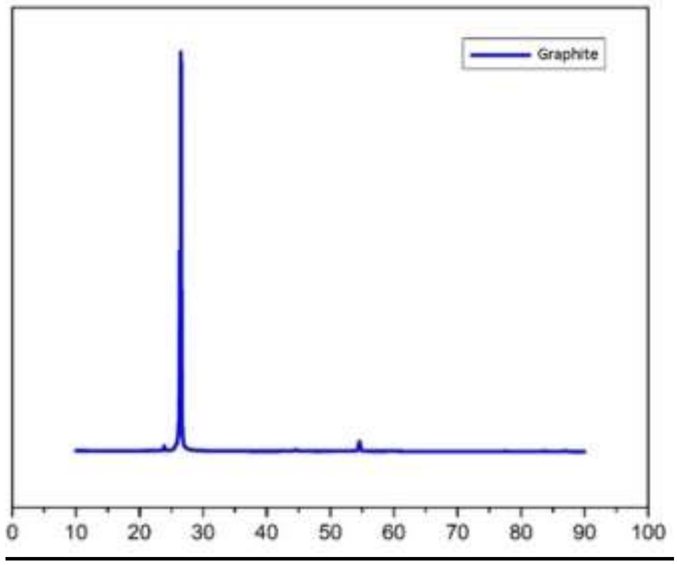

Figure 1. Graph of the relationship of diffraction (2θ°) to intensity (Cps) on the result XRD Graphite

The XRD results shown in Figure 1 show that the graphite powder has a single phase, namely graphite with a composition of 100% and have a hexagonal crystal structure.

3.2 Mortar Flexural Strength Test

3.2.1 Cyclic Voltammetry (CV) Testing Analysis

Cyclic Voltammetry (CV) is a basic electrochemical test for materials. In this case, the current is recorded by sweeping alternating potentials (from positive to negative and negative to positive) between the selected boundaries [8]. The resulting voltamogram is a curve between current (on the vertical axis) versus potential (on the horizontal axis).

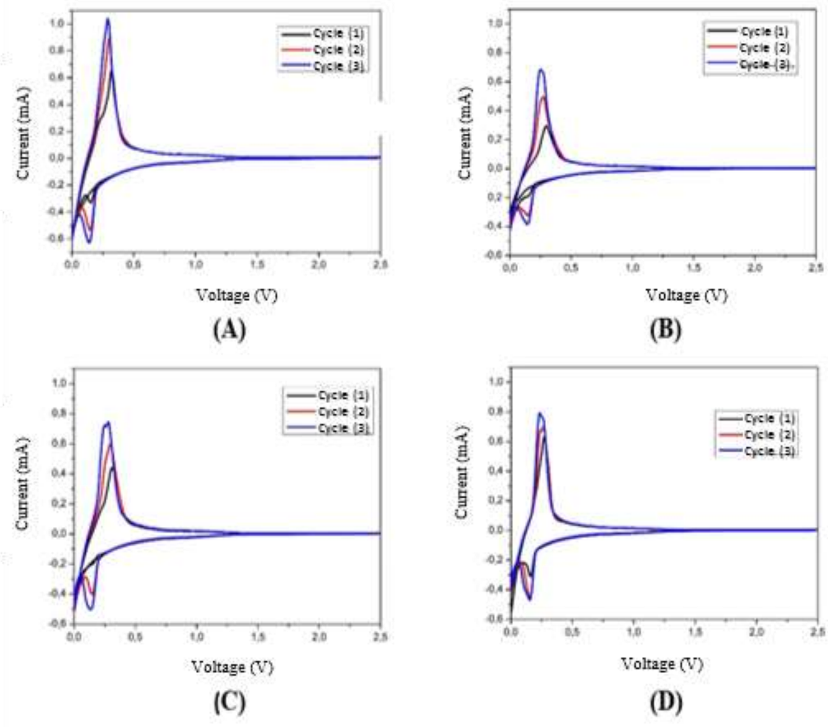

Figure 2. Cyclic voltamogram on samples (A) 17.85%(0.1 mm), (B) 20.8% (0.1mm), (C)25% (0.07 mm), (D) 25% (0.1 mm))

The CV curves of the four samples showed very similar voltage-current performance and were tested at a scan rate of 0.1 mV/s for three cycles. The main redox peak currents occurred in samples with a solid content of 17.8% from cycle one to cycle three of 0.13 V, 0.15 V and 0.13 V for reduction during cathodic scanning, and about 0.30 V, 0.26 V, and 0.24 V for oxidation during scanning. anodic. The distance between the peaks of the redox curve of sample A is calculated at 0.17 V, V, 0.11 V and 0.11 V.

The graph in the four samples above from three cycles basically has a reduction peak of around 0.4-0.8 V in the first cycle. However, these peaks disappeared in the second and third cycles, indicating that the formation of solid electrolyte interphase (SEI) could further suppress electrolyte decomposition effectively after the first cycle.

3.3 Charge-Discharge Testing Analysis (CD)

The dynamics of charging and discharging a battery can be characterized by measuring the voltage with a constant charge and current input discharge [9]. Testing of battery cells is carried out with a charge/discharge process to obtain battery cell performance so that a large battery cell capacity is obtained. In the charge/discharge test, all graphite anode samples underwent a charge and discharge process so that this graphite anode had met the initial requirements to be applied as a secondary battery.

During the discharge process, an oxidation reaction occurs at the LiC6 anode which satisfies the reaction equation:

$$LiC6 \rightarrow Li+ + C6 \quad (1)$$

At the time of charge a reduction reaction occurs with the reaction equation:

$$Li+ + C6 \rightarrow LiC6 \quad (2)$$

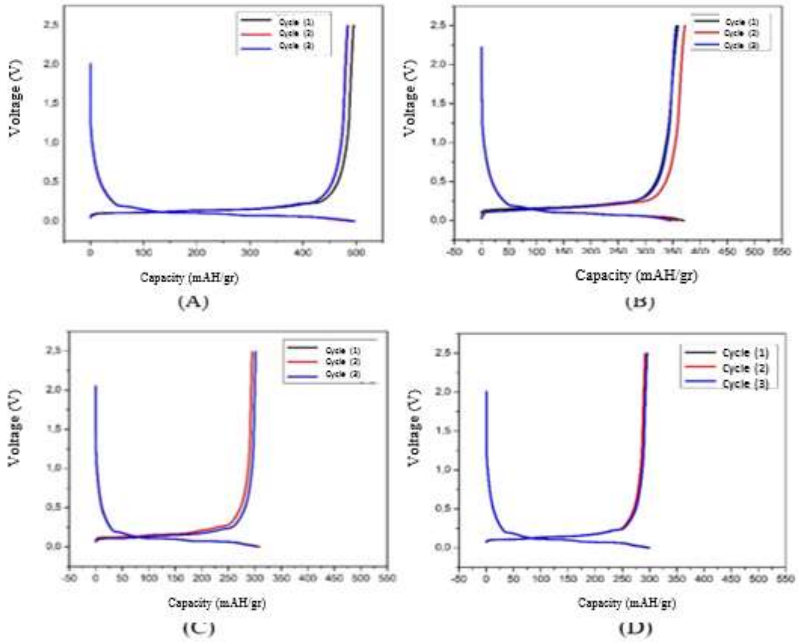

Figure 3. Graphite anode charge and discharge curve (A) 17.85% (0.1 mm), (B) 20.8 (0.1 mm), (C) 25% (0.07 mm), (D) 25% (0.1mm)

Figure shows that there have been three charge-discharge cycles in the sample A to sample D batteries. The graphite discharge curve shows several voltage plateaus corresponding to the formation of lithium-graphite intercalation compounds and Li^+ deintercalation plateaus. In samples B to sample D no significant differences were observed in the charge and discharge voltage levels of these samples. In the first charge cycle, the battery is empty and starts with a voltage of 2.4 V. Meanwhile, sample A with the smallest solid content of 17.8% has a larger charge discharge capacity than the other samples. This condition indicates that the total charge obtained in the discharge condition in sample A is more and better than the other samples.

3.4 Electrochemical Impedance Spectroscopy (EIS) Testing Analysis

Electrochemical impedance spectroscopy (EIS) is a very effective methodology for determining the electrochemical and impedance behavior of lithium batteries without cell damage. So that the EIS data can be thoroughly analyzed using an equivalent circuit model consisting of circuit elements, such as resistors, capacitances, Warburg impedance (Wz), and constant phase elements (CPE). If a suitable equivalent circuit model is selected, the calculated parameters of the circuit elements provide a comprehensive understanding of the electrochemical and physical phenomena in the cell [10].

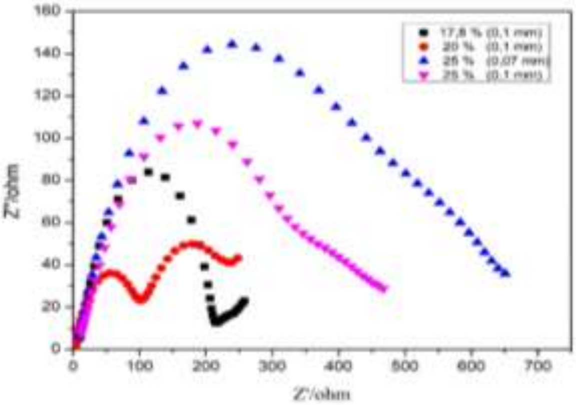

Figure 4. Graphite anode EIS graph on samples (A) 17.85% (0.1 mm), (B) 20.8% (0.1 mm), (C) 25% (0.07 mm), (D) 25% (0.1 mm)

This pattern can be used as a reference for the ability of the electrode to store lithium ions for use in lithium-ion batteries. The narrower the pattern, the higher the conductivity value. This is because the total resistance (impedance) is getting smaller along with the narrower the semicircle pattern.

4 Conclusion

The slurry obtained from mixing Graphite powder, PVDF, Super-P and various DMAC solvents has been successfully prepared with solid content in samples A: 17.8% (0.1mm), B: 20.8% (0.1 mm), C: 25% (0.07 mm) and D: 25% (0.1 mm). The best anode sheet was obtained by using 2.5 mL of DMAC solvent with a solid content of 25% at an anode sheet thickness of 0.07 mm. The results of the battery performance test show that the thicker the sheet, the smaller the capacity. A battery with 25% solid content at 0.1 mm thickness has a smaller capacity of 296.46 mAh/g in the first cycle. As for the battery with a solid content of 25% at a thickness of 0.07 mm, it has a capacity of 309.33 mAh/g in the first cycle.

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