The Effect of Activated Carb on Derived from Black Betel Leaf Biomass Waste as Composite Anodes on Lithium-Ion Battery Applications

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Abstract. Lithium-ion batteries have shown promising performance in high-energy storage systems for electric vehicles. The electrode material used in the battery affects the performance of the LIB. The material on the anode can be modified by adding activated carbon (AC) to the graphite. AC can be made from a variety of biomass wastes, including black betel leaf biomass. AC was prepared by hydrothermal carbonization method in an inert gas atmosphere and then activated with a KOH solution. AC material was then analyzed by SEM and FTIR. Li-ion batteries with 0%, 10%, and 20% activated carbon addition were tested with a battery analyzer. The resulting specific capacities of graphite-AC 0%, graphite-AC 10%, and graphite-AC 20% batteries were 115.57 mAh/g, 94.60 mAh/g, and 76.38 mAh/g, respectively. The battery was then cycle tested at a current of 0.5C, and the resulting battery with the addition of 20% activated carbon showed the best retention capacity of 88.34% after 50 cycles. The battery test results show that activated carbon from black betel leaves can be used as an anode material for lithium-ion batteries.

1 Introduction

Developing the economy, technology, and sciences demands the action of renewable and sustainable energy. Lithium-ion batteries are known as one type of electrochemical energy device that is efficient, reliable, and practical for the storage and conversion of electrical energy generated so that it can be used in various applications [1]. LIB has the advantage of being a clean and sustainable energy source, such as solar, wind, waterfall, and geothermal energy sources [2]. One way to improve the performance of lithium-ion batteries, such as developing lithium-ion battery electrode materials, especially anode materials. Some research conducted in Indonesia related to lithium-ion batteries focuses on making active materials that make up the battery [3]. Addition of materials such as graphite [4], graphene, graphene oxide [5], carbon nanotubes, black carbon, soft or hard carbon [6], metal, and metal oxide [7] can improve the electrochemical parameters and properties of the battery anode. Therefore, in order to increase the lithium-ion battery's performance, activated carbon is added to the graphite anode.

Activated carbon has been extensively utilized as an electrode material for several devices because of its inexpensive cost, good electrical conductivity, high specific surface area, and chemical resistance [8]. Activated carbon is made from various natural materials such as coir and coconut shells [9], mango waste [10], corn cobs, banana peel [11], chicken bones, cow bones, and fish bones [12]. Activated carbon used as a lithium anode material aims to expand the material's surface with the appearance of pores in the material and can increase its absorption properties [13].

The utilization of activated carbon as electrodes and supercapacitors has been widely carried out. Activated carbon from biomass waste can also be utilized as a raw material for air cathodes. The highest voltage is obtained in NaOH electrolyte with an anode in the form of aluminium and activated carbon derived from empty palm bunches [14]. In 2020, Pahlevi et al. tried to make an activated carbon-based battery prototype from bamboo activated with KOH and tested with NaOH.
electricity, producing 103.03 watts of power [15]. Activated carbon utilized as electrodes and supercapacitors, will help solve problems in biomass waste.

In this study, activated carbon derived from black betel leaf biomass is added to graphite anodes, which aims to increase the capacity and cycle life of lithium-ion batteries. The battery is assembled on a cylinder cell type 18650 with an LNMC 811 cathode. The battery is tested using a battery analyzer to establish the impact of the addition of activated carbon to the graphite anode on battery performance.

2 Experimental Methods

2.1 Material Preparation

The preparation of activated carbon from black betel leaf biomass was made by drying the biomass for 1 hour. The drying results were then carbonized with the HTC (Hydrothermal Carbonization) process at 250°C for 2 hours under inert conditions using nitrogen gas [16]. After the carbonization process, the carbon is then chemically activated with the help of a chemical compound solution such as KOH with an activator-carbon ratio of 3:1 [17]. Then soaked the carbon for 24 hours to ensure the continuity of the diffusion process in the carbon pores. Carbon that has passed the chemical activation process is then physically activated at 800 °C for 60 minutes. The physical activation process on carbon will remove volatile impurities, enlarge the cavity structure, and remove impure hydrocarbons on carbon [18]. The final stage of this process is washing the charcoal with water until the pH is neutral. The activated carbon is then dried at 105 °C to remove moisture trapped in the pores [19]. The activated carbon material was then tested for SEM characterization to identify the material's morphology and by FTIR for its functional groups.

2.2 Material Characterization

Carbon and activated carbon from black betel leaf biomass were characterized using a scanning electron microscope (SEM) (JEOL JSM-6510LA, Japan) to examine the morphs of the samples. A Shimadzu (Japan) FTIR spectrometer with a mid-IR region (4000-400/cm) was used to analyze the functional groups of the materials.

2.3 Electrochemical Measurement

Figure 1 illustrates the method of creating anode electrodes from graphite and black betel-activated carbon. Adding the activated carbon to graphite anodes with a percentage of 0%, 10%, and 20%. In the manufacture of anodes, it is necessary to add some materials, such as carboxymethyl cellulose (CMC) as an adhesive, styrene-butadiene rubber (SBR) as a binder, acetylene black (AB) as a conductive material, and water (H2O) as a solvent.

Fig. 1. Anode manufacturing scheme

The materials were mixed and stirred using a motorized stirrer to a homogeneous slurry. The resulting slurry was coated using a coater tool on copper foil with a thickness of 200 microns. The coating was dried in an oven at 90°C over night. The electrodes were then assembled in an 18650-type cell cylinder.

3 Result and Discussion

3.1 The Properties and Discussion

Table 1 The Properties Of Activated Carbon

<table>
<thead>
<tr>
<th>No</th>
<th>Properties</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Yield</td>
<td>2.33%</td>
</tr>
<tr>
<td>2</td>
<td>Water content</td>
<td>14.75%</td>
</tr>
<tr>
<td>3</td>
<td>Carbon content</td>
<td>21.33%</td>
</tr>
<tr>
<td>4</td>
<td>Activated carbon content</td>
<td>20.01%</td>
</tr>
</tbody>
</table>

Table 1 presents the analytical results of the activated carbon produced. The yield of activated carbon from black betel leaves is 2.33%; that is, 6 grams of activated carbon are produced from 300 grams of processed biomass. This low yield is due to the low cellulose content of black betel leaves, which is small (2–3%) [20], compared to the cellulose content of various types of wood, whose levels are more than 50%. According to Lu (2017), the main factor determining the production of activated carbon and the mechanism of chemical activation is the cellulose content of the material [16]. Determining of water content aims to determine the hygroscopic nature of activated carbon. The moisture content contained in activated carbon is 14.75%. The moisture content of activated carbon still fulfills the standard limit of SNI 06-3730-1995 on technical activated carbon, where the moisture content in activated carbon is a maximum of 15%. High water content will reduce the quality of activated carbon because water adsorbed on activated carbon will reduce the capacity and adsorption capacity of liquids and gases [14].

Then the carbon and activated carbon were burned at 900 °C in a furnace to determine the pure carbon content contained in the resulting activated carbon. The non-activated carbon has a carbon content of 21.33%, while the activated carbon content after activation is 20.01%. KOH activator dissolves minerals bound to
carbon molecules and replaces them with a functional group [15].

3.2 Material Characterization

The results of the FTIR test on carbon and activated carbon showed the identification of O-H, C=O, C=O, and C-O functional groups. The foundation of these findings is the correspondence between the appropriateness of functional groups at specific absorbance peaks and wave number areas found in the literature [21], which can be seen in Figure 2. The spectra of activated carbon are slightly different from those of carbon without activation. The change in spectra shape, shift, intensity reduction, and addition of new peaks after the activation process is likely due to the carbonization and activation processes causing dehydration and the decomposition of complex lignocellulose groups into more straightforward groups [22].

![FTIR test result of carbon and activated carbon](image1)

**Fig. 2.** FTIR test result of carbon and activated carbon

At wavelengths between 3400 and 3600 cm\(^{-1}\) there is a broad and sloping peak on carbon and activated carbon, which indicates the presence of O-H functional groups from hydroxyl groups with hydrogen bonds. This O-H bond may come from hydrogen bonds, so it is possible to interact with water molecules adsorbed by activated carbon samples [23]. At wave number 2373 - 2314 cm\(^{-1}\) there is a C triple functional groups from stretching alkyne groups. This is following the research of Ahmad et al. (2014) and Rattanapan et al. (2017), who found that the O-H functional group ranges from 3200-3700 cm\(^{-1}\) and the C triple is at wavelengths 2067-2900 cm\(^{-1}\) [24],[25].

The C=C functional group with C=O has a similar wavelength location; to distinguish the C=C active group contained at a wavelength of 1542 cm\(^{-1}\), it is an active group on activated carbon derived from plants [26]. While the C=O active group found between 1800 and 1650 cm\(^{-1}\) wave numbers is an active group on activated carbon derived from animals [12]. These wave numbers are not much different from the results of research by Ahmed et al. (2014), which show that the aromatic ring C=C group is shown at wave numbers 1400–1583 cm\(^{-1}\). Then, for the wave numbers 1073 cm\(^{-1}\) on carbon and 1004 cm\(^{-1}\) on activated carbon, the C-O stretching functional groups of ethers, esters, or phenols [24],[25]. Additionally, this is in line with the findings of Doke and Khan's (2017) study, which indicated that the C-O functional groups of ethers and esters manifest at wave numbers between 1060 and 1319 cm\(^{-1}\) [27].

The characterization of the morphology and size of the samples was carried out by SEM (Scanning Electron Microscope). Carbon and activated carbon samples were SEM tested with 1000 times magnification. The morphology of the carbon sample is well identified, and each grain has an average dimension ranging from 5.96 µm, which can be seen in Figure 3(a). From the picture, the carbon has tiny pore holes. In contrast, activated carbon has formed irregular and heterogeneous pores with average dimensions ranging from 2.91 µm, which can be seen in Figure 3 (b). On activated carbon, the sample’s morphology is identified, and aggregation is seen. Aggregation occurs due to an increase in the physical properties of the material particles, such as temperature. In general, lignin can maintain the morphological structure of biomass because it contains esters and ethers with cross-links, after the impregnation and activation process with KOH, the lignin structure and cell walls are destroyed so that the pores formed from activated carbon do not have a typical shape or are similar to the morphological structure of the raw material [16].

![SEM test result on carbon sample (a) and activated carbon (b) of black betel leaf with 1000 times magnification](image2)

**Fig. 3.** SEM test result on carbon sample (a) and activated carbon (b) of black betel leaf with 1000 times magnification

Pore formation, or expansion into larger pores, is caused by the reduction of impurities on the carbon surface [22]. The interaction between KOH as an activator agent and carbon also increases the reaction rate when the precursor is heated at a high temperature (800°C) to cause pore development and pore formation [24]. The construction of micropores is generally due to the accumulation of volatile substances on the particles and their splitting. When secondary splitting occurs with increasing reaction temperature, flammable substances will be released from the particles and form carbon that is deposited so that it blocks the establishment of pores, which will reduce the surface area of micropores and then cause micropores to break and merge into bigger pores, increasing the external surface of the pore [28]. The shapes of the pores in activated carbon show that impurities or activator compounds have not entirely vanished from them, which means that the absorption of the activated carbon is not at its most effective.
3.3 Electrochemical Characterization

The graphite anode material was mixed with activated carbon derived from black betel leaves at 0%, 10%, and 20%. The battery is assembled in the form of cylindrical cells of type 18650 with an LNMC 811 cathode, Celgard separator, and LiPF6 electrolyte in EC: EMC 3: 7. The results of testing using a battery analyzer show a comparison of the specific capacity of the three variations of activated carbon addition to the battery, as seen in Figure 4. The specific capacities produced by Graphite-AC 0%, Graphite-AC 10%, and Graphite-AC 20% batteries are 115.57 mAh/g, 94.60 mAh/g, and 76.38 mAh/g, respectively. Of the three samples, the one with the addition of 20% has the smallest specific capacity compared to the other samples. These results have an unfavourable value because, in general, with increasing loading, there is a decrease in voltage and capacity in the battery due to resistance in the battery. The cause of resistance in the battery is the less conductive battery sheet constituent materials, such as PVDF and electrolyte. A reduction in specific capacity also comes on by poor contact between the cathode and anode sheets on the current collection sheet.

![Graph showing specific capacity of lithium-ion battery with variation of activated carbon material](image1)

Fig. 4. The specific capacity of lithium-ion battery with variation of activated carbon material

The graphite anode material was mixed with activated carbon derived from black betel leaves at 0%, 10%, and 20%. The battery was then cycle tested at a charge current of 0.5C and a discharge current of 1C to determine the life cycle of the battery. From the test results, it was found that the best retention capacity, namely 88.34% after 50 cycles. This shows that activated carbon from black betel leaf can be used as lithium-ion battery anode material.

![Graph showing cycle life of lithium-ion battery with variation of activated carbon material](image2)

Fig. 5. The cycle life of lithium-ion battery with variation of activated carbon material

These results showed that activated carbon from black betel leaves can be used as an additional active material on graphite anodes. However, it still needs to be developed again to conductivity testing of activated carbon materials to determine the effect of material conductivity on battery performance.

4 Conclusion

Activated carbon from black betel leaves with a KOH activator has been successfully made by the hydrothermal carbonization method, and the results have complied with the proximate analysis standard for activated carbon according to SNI 06-3730-1995. Characterization of activated carbon using FTIR showed that the functional groups detected on activated carbon were O-H, C≡C, C=C, and C-O groups. The samples showed non-uniform particle distribution and aggregation. The morphology of the activated carbon material has dimensions ranging from 2.91µm. Activated carbon is added to graphite anodes with percentages of 0%, 10%, and 20%. The battery was assembled in the form of a cylindrical cell type 18650 with an LNMC 811 cathode, Celgard separator, and LiPF6 electrolyte in EC : EMC (3 : 7). Li-ion batteries with the addition of 0%, 10%, and 20% activated carbon were tested with a battery analyzer, and the specific capacities produced by Graphite-AC 0%, Graphite-AC 10%, and Graphite-AC 20% batteries are 115.57 mAh/g, 94.60 mAh/g, and 76.38 mAh/g, respectively. The battery was then cycle tested at a charge current of 0.5C and a discharge current of 1C to determine the life cycle of the battery. From the test results, it was found that the battery with the addition of 20% activated carbon showed the best retention capacity, namely 88.34% after 50 cycles. This shows that activated carbon from black betel leaf can be used as lithium-ion battery anode material.

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